The Stratospheric Aerosol Layer during 1964 and 1965*

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ABSTRACT

Observations of stratospheric aerosols have been made with an optical radar at Lexington, Massachusetts, during a two-year study. Some observations were also conducted at College, Alaska, in the summer of 1964. Vertical profiles of aerosol concentration were obtained by comparing the optical radar echoes with the expected return from a molecular atmosphere; the observed signal from 25-30 km altitude was used to calibrate the instrument.

The observations show that the aerosol layer near 20 km exhibited little temporal variability. The observed return from the layer was approximately 1.9 times the return from a molecular atmosphere; the daily rms fluctuations were smaller. Comparisons with previous measurements indicate that the concentration of stratospheric aerosols was one order of magnitude higher, presumably because of the eruption of Mount Agung, in March 1963.

The data have been compared with various meteorological parameters associated with conditions in the lower stratosphere. A significant negative correlation between fluctuations of dust and ozone has been found in the measurements.

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1. INTRODUCTION

The presence of a layer of particulate material in the lower stratosphere was deduced many years ago from twilight observations of the purple light (Gruner and Kleinert, 1927; Gruner, 1942).

The existence of the layer was directly verified with particle sampling equipment carried on high-altitude balloons (Junge, Chagnon, and Manson, 1961; Chagnon and Junge, 1961). Vertical profiles of particle concentration obtained with an inertial impactor designed to collect particles larger than ~ 0.1-micron radius persistently exhibited a broad maximum in particle concentration at ~ 20-km altitude. Collections by impactors on U-2 aircraft at 20 km (Friend et al., 1961; Junge and Manson, 1961) showed that particle concentrations between 60°S and 70°N exhibited no systematic latitudinal variation, and the aerosol layer was a world-wide feature of the lower stratosphere.

Chemical analyses of the particles collected by the impactors indicated that the majority of the particles were composed of sulfates (Junge, Chagnon, and Manson, 1961). This result was confirmed by electron-diffraction studies of the U-2 samples; Friend et al. (1961) found that more than 90% of the mass of the impacted particles was composed of crystalline ammonium sulfate or, on occasion, ammonium persulfate. The chemical composition of the particles and the observed shape of the concentration profiles led Junge to propose that the particles were formed in situ by oxidation of traces of gaseous sulfur compounds (hydrogen sulfide and sulfur dioxide) that had entered the stratosphere by vertical mixing processes.

The presence of the aerosol layer has been substantiated by other investigators. For example, Bigg (1956) performed photometric observations of the rate of change of the intensity of twilight and related rapid variations of this parameter to the interaction of the edge of the earth's shadow with regions of dust particles in the atmosphere. These measurements have detected the presence of appreciable amounts of dust in the 15-20 km region, thereby supporting the results of the particle

collections, although the concept of a well-defined illumination discontinuity at the shadow boundary has been criticized, particularly by Megrelishvilli (1958).

Volz and Goody (1962) measured the absolute intensity of the twilight at a fixed angle of elevation to derive quantitative information on the turbidity of the upper atmosphere. On the basis of a carefully constructed theoretical model which incorporated the effects of scattering by the molecular and aerosol components of the atmosphere and absorption by atmospheric ozone, dust profiles were derived by comparing the observed twilight intensity with the expected intensity from the model atmosphere. Even though these results provide data that are smooth over at least 10-km altitude, the turbidity profiles usually displayed a broad maximum between 15 km and 30 km in accordance with a stratospheric dust layer. A comprehensive treatment covering observations and theory of twilight scattering phenomena is given by Rosenberg (1966).

Observations obtained by other techniques have also corroborated the existence of the layer and, in some cases, provided additional details about it. An observation of the vertical distribution of the diffuse radiation from the daytime sky measured by rocket photometry (Rössler, 1963) showed a radiation maximum at ~ 25 km that could be attributed to aerosol scattering. This rocket sounding, obtained in the Sahara, substantiates the measurements reported by Chagnon and Junge (1961) which indicated that the concentration maximum may be located at higher levels in tropical regions.

The aureole photometry of Newkirk and Eddy (1964) also demonstrated the presence of the layer. The angular and spectral distribution of sunlight scattered in the stratosphere measured by a balloon-borne coronagraph was analyzed to provide information on the particulate material above the instrument and, by differentiation, on local aerosol parameters. The dust concentrations observed on two different flights of the instrument were consistent with the direct-sampling results. Evidence was also obtained during this study that the dust "layer" may contain thin laminae with concentrations exceeding smoothed values by at least an order of magnitude. Such stratifications were also detected, at times, by the twilight observations of Bigg (1964).

On occasion, thin dust layers in the lower stratosphere can be observed visually and photographically when viewed in the horizontal direction from aircraft or balloons (see, for example, Jacobs, 1954; Bull and James, 1956; Ross, 1958); these observations, however, could usually be associated with preceding volcanic eruptions and may not be a natural feature of the strospheric aerosol distribution.

Searchlights have been used to detect the presence of scattering layers in the atmosphere (Rosenberg, 1960; Elterman and Campbell, 1964). With this technique a searchlight beam directed into the atmosphere is scanned photometrically to measure altitude variations of the light scattered by atmospheric constituents. These observations have also shown strong aerosol scattering near 20 km. Recent research in the Soviet Union (Rosenberg, Sandomirsky, and Poldmaa, 1966) using twilight photometry and measurements from aircraft and spaceships (Rosenberg and Nikolaeva-Tereshkova, 1965), as well as searchlight observations, has consistently detected the aerosol layer at 15-22 km altitudes, with minimum dust amounts observed in the 25-30 km region.

The eruption of Mount Agung on Bali (8°25'S, 115°30'E), on 17 March 1963, has increased the aerosol content of the stratosphere. Enhanced optical effects have been reported by many observers: Volz (1965) has recently summarized many of the visual and photometric observations obtained since the volcanic eruption. In the Northern Hemisphere reports of unusual twilight effects (see, for example, Meinel and Meinel, 1963 and 1964; Volz, 1964) indicate that the dust content of the stratosphere increased substantially during the fall of 1963 and that abnormal conditions have persisted at least throughout 1964.

Some quantitative measurements of the increase of stratospheric aerosol concentrations before and after the Agung eruption with the same method of observation used are available. Volz (1964, 1965) has continued earlier measurements of the intensity of the twilight (Volz and Goody, 1962). These observations show deviations from normal conditions, beginning as early as May 1963; a pronounced increase in stratospheric turbidity early in the winter of 1963 was followed by abnormally high

dust amounts since that time. Volz has estimated that if enhanced turbidity was due to a layer that is only a few kilometers thick, the aerosol concentration may have exceeded that of the ordinary sulfate layer by a factor of 20 during the winter 1963/64.

Recent studies of the aerosol layer using balloon-borne photoelectric particle counters (Rosen, 1964) also indicate that particle concentrations between 15 km and 20 km may be an order of magnitude larger than the results of the particle collections of Junge and his collaborators. A quantitative comparison between the results of optical measurements and the particle concentrations obtained by analyzing impactor samples is, however, subject to some question in view of the uncertainties associated with the collection efficiency of an impaction surface and the possible loss of volatile material after collection.

Stratospheric particles have been collected with impactors both before and after the Agung event and, therefore, provide comparative information about the perturbed state of the aerosol layer. Particle-number concentrations measured by Friend (Feely et al., 1963) from U-2 flights at latitudes near 30°N, on 7 May 1963 and 30 July 1963, were approximately four times greater than values measured before the volcanic eruption. This was attributed to natural variations in the ordinary sulfate layer associated with the existence of a laminar structure. It now seems possible, however, that these measurements may have recorded the incursion of volcanic material into the Northern Hemisphere with little delay from events in the Southern Hemisphere.

Particle collections on impaction surfaces mounted on U-2 aircraft have also been performed in the Southern Hemisphere before and after the Mount Agung eruption by Mossop (1963, 1964). Aerosol concentrations up to 7 times those observed before the eruption were found in the impactor samples for almost a year after the eruption. Furthermore, the dimensions of many of the particles were much larger than observed in samples collected before the eruption. This was due not only to the presence of larger particles which could often be recognized as volcanic debris, but also to the existence of a layer of water-soluble material on

these particles and, on occasion, of a fluid layer surrounding the particle. Mossop suggested that the coating could be sulfate material or, in the case of fluid layers, sulfuric acid resulting from oxidation of the large quantities of gaseous sulfur compounds injected into the stratosphere by the eruption. It is possible that the pronounced changes in the optical properties of the aerosol layer may be related to changes in the physical and chemical properties of the stratospheric aerosol, in addition to enhanced effects resulting from increased particle concentrations.

The present study stems from exploratory attempts to provide routine observations of atmospheric aerosols by using a pulsed ruby laser as the source of radiation for an optical radar system. The apparatus developed for the study is described here.

The use of light beams to derive information about the upper atmosphere is not a new concept; the ability to detect aerosol layers in the atmosphere by measuring light scattered from a searchlight beam has already been discussed. The use of searchlights was originally suggested by Synge (1930) as a means of measuring atmospheric densities; several investigators have since used searchlight beams to obtain atmospheric density profiles (see, for example, Elterman, 1954). Searchlight observations are usually performed by photometrically scanning a steady or modulated searchlight beam by photographic or electronic techniques from an observing site located a considerable distance from the source, and employing triangulation for altitude determination. A pulsed searchlight, capable of being operated from a single site by using the time elapsed between the transmitted and received signal for altitude determination was used by Friedland, Katzenstein, and Zatzick (1956); however, suitable light sources were not yet available to take full advantage of a pulsed system.

2. INSTRUMENTATION

The equipment used for the detection of stratospheric aerosols is an optical radar; a pulsed ruby laser acts as the transmitter and an astronomical telescope as the receiver for a monostatic radar system. Previous applications of this technique to the study of the atmosphere have been presented in an earlier note by Fiocco and Smullin (1963), and in subsequent papers.

The details of the instrumentation have changed throughout this study. The apparatus utilized in the early phase of this work and until the end of May 1964 has been described and some initial results have been reported by Fiocco and Grams (1964). During the summer of 1964, two optical radars were taken to Alaska and Sweden to study noctilucent clouds (Fiocco and Grams, 1966); some data on stratospheric aerosols which were collected in Alaska with one of these instruments are presented here. From the fall of 1964 to the end of the study, data were collected in Massachusetts with the apparatus previously located in Sweden. In the present version the apparatus utilizes a Cassegrainian receiving telescope of 40-cm diameter. The receiver includes a 6 Å bandwidth interference filter and an EMI 9558A photomultiplier that can be refrigerated by circulation of methanol cooled by mixing with dry ice.

The laser unit, built by Applied Lasers, Inc., is capable of delivering pulses of approximately 2 joules, with a duration shorter than 1 μsec at a maximum pulse repetition rate of approximately 30 per minute. The ruby rod, flashlamp (EG & G FX-67 A), and the elliptical cavity are cooled by closed-loop circulation of distilled water. The 90° orientation ruby rod is 6 5/8 inches long and 3/8 in diameter; a rotating prism is used for Q-switching the laser. In the first phase of this work we used a ruby laser, built by the Radio Corporation of America, which could emit pulses of approximately 1/2 joule and duration shorter than 1 μsec. Q-switching was achieved through the use of a rotating mirror. The 3-inch ruby rod used in this laser and the elliptical cavity were cooled by dry nitrogen passed through a coil immersed in liquid nitrogen. An EG & G FX-42 flashlamp was modified for cooling with distilled water.

The radiation is collimated by a transmitting telescope, 10 cm in diameter with 1-meter focal length. The instrument includes two synchronized rotating shutters, one for cutting out any fluorescence emitted by the laser unit after the main pulse is radiated and another synchronized shutter to prevent exposing the photomultiplier to the intense return obtained from scattering at short distances.

3. DATA REDUCTION

When the laser is pulsed, the return signals are displayed on a Tektronix 555 dual-beam oscilloscope and photographed. Two traces displaying the amplified photomultiplier current with different sweep rates are recorded simultaneously: one trace displays echoes at 0-40 km altitudes, while the other extends the observation to a range ≈ 200 km. At altitudes above 30 km, the flux density of the echoes decreases to such an extent that a continuous current is no longer recorded and, when the gain is increased by a factor of ~ 100, the display consists of "spikes" representing the emission of single photoelectrons by the photocathode. The 200-km data can be analyzed by counting the number of "spikes" observed in specified altitude intervals for a large number of traces (Fiocco and Smullin, 1963).

Most of the 1964 data were recorded with a Hewlett-Packard Model 196A oscilloscope camera capable of recording several consecutive optical radar echoes on Polaroid film, and were digitized by use of a calibrated grid. From January 1965 to the end of the study, data were recorded on 35-mm film with a modified Fairchild 556-BH1 radarscope camera. The reduction of these data has been accomplished with a Benson-Lehner "Oscar F" semiautomatic record analyzer. The observed coordinates for each 40-km display were digitized and punched on data-processing cards for subsequent analysis on the IBM 7094 computer. In either case, corrections for any nonlinearity of the oscilloscope display or camera system were included.

Since the pulse length of the laser is less than 1 μsec, a range resolution of at least 150 meters is available in the oscilloscope records. In view of the presence of random components in the received signal, however, it was found necessary to integrate a number of successive traces and obtain an average optical radar return for the given time interval. The number of traces required to reduce the statistical fluctuations of the received signal to an acceptable level, the relatively large range interval used for the study, and the difficulty of determining the exact position of very small range intervals, especially on the Polaroid prints,

made it desirable to decrease the range resolution. The data records were digitized by using average vertical coordinates for 1-km altitude increments for the Polaroid data and 0.5-km increments for the data recorded on microfilm. The observed coordinates for consecutive traces were summed to provide an integrated optical radar return for each period of observation. This average signal was then compared with the expected signal from a molecular atmosphere to construct vertical profiles of the dust content of the stratosphere.

4. OPTICAL RADAR EQUATIONS

The intensity of echoes for an optically thin layer of homogeneously distributed scatters, expressed as $\frac{dn_r}{dR}$, the expected number of photoelectrons per transmitted pulse per unit range, emitted at the photomultiplier cathode, is

$$\frac{dn_r}{dR} = \frac{n\lambda}{hc} W_t A_r k_r k_t k_a^2 \frac{\Sigma}{4\pi R^2} \tag{1}$$

where W_t is the transmitted energy per pulse, A_r is the collecting area of the receiving telescope, k_r and k_t are the efficiencies of the receiving and transmitting systems, k_a is the atmospheric transmission, Σ is the collective radar cross section of the scatterers per unit of volume, R is the distance, λ is the wavelength, n is the quantum efficiency of the photocathode, h is Planck's constant, and c is the speed of light.

Σ is determined by contributions from various atmospheric constituents. We shall interpret our returns according to a model that assumes that the stratosphere is composed of air molecules and dust particles. The radar scattering cross section of air molecules will be calculated with the aid of the expression

$$\Sigma_M = N_M \sigma_M = \frac{16\pi^3}{\lambda 4} \frac{(\tilde{n}-1)^2}{N_M}, \tag{2}$$

where $\tilde{n}$ is the refractive index of air, N_M is the molecular number density, and σ_M is the radar cross section of an individual molecule.

We shall assume that the dust particles are homogeneous spheres of radius r and describe the size distribution with the aid of a commonly used parameter

$$\xi(r) = dN/d \log r, \tag{3}$$

where N is the concentration of dust particles of radius smaller than r. N_D, the number of dust particles in a radius interval from r_1 to r_2, is thereby obtained from the integral

$$N_D = \int_{r_1}^{r_2} \xi(r)\, d \log r = 0.434 \int_{r_1}^{r_2} \xi(r) \frac{dr}{r}. \qquad (4)$$

When the size distribution function is specified, the results of the Mie scattering theory (van de Hulst, 1957) can be used to evaluate the radar cross section for the ensemble of dust particles. The size distribution obtained by analyses of the impactor samples of stratospheric particles collected by Junge et al. (1961) and Friend et al. (1961) showed a regular decrease in concentration for particles in the size range from approximately 0.1 μ to at least 1 μ radius that could be approximated by a power law:

$$\xi(r) = \frac{C}{r^{\nu}}, \qquad (5)$$

where C and ν are constants. In a subsequent investigation Friend (Feely et al. 1963) found the lower radius limit for the power-law approximation at a slightly larger particle size; the distribution function was approximately symmetric about 0.275 μ radius. It should be pointed out that the determination of radius limits for the observed size distribution is experimentally difficult. At the lower limit, direct sampling measurements require corrections for impaction efficiency, and optical measurements are rather insensitive to changes in the size distribution. At the upper limit the observed low particle concentrations are subject to large statistical fluctuations. Accepted values for the exponent ν range between 3 and 4. Junge (1963) uses Friend's value, $\nu = 3.5$, which is also consistent with the optical measurements of Newkirk and Eddy (1964).

We have evaluated Mie intensity functions for backscattering from homogeneous spheres at complex values of the index of refraction to compute the radar cross section for an individual dust particle, σ_D, as a function of its size. Complete results, which we hope supplement the extensive literature on the subject, will be presented elsewhere.

We have limited our present work to an analysis of the scattering cross section for nonabsorbing spheres with a refractive index $\tilde{n}=1.5$. This

value is in accordance with the composition of the stratospheric particles collected by Junge et al. (1961) and Friend et al. (1961), and it has also been used by Volz (1954) in a study of atmospheric aerosols.

Fig. 1 shows the normalized radar cross section $\sigma/\pi r^2$ as a function of the parameter $a = 2\pi r/\lambda$ for $\tilde{n} = 1.5$; calculations were performed in steps $\Delta a = 0.05$ to a value $a = 30$. The scattering function has been integrated for the following model of size distribution.

$$\xi(r) = \begin{cases} \dfrac{C}{r^{\nu}} & \text{for } r_1 \leq r \leq r_2 \\ 0 & \text{for } r < r_1 \text{ or } r > r_2. \end{cases} \tag{6}$$

Thus

$$N_D = \frac{0.434C}{\nu}\left(\frac{1}{r_1^{\nu}} - \frac{1}{r_2^{\nu}}\right) \approx \frac{0.434C}{\nu r^2} \tag{7}$$

for $r_2 \gg r_1$, and the radar cross section for the ensemble of particles is

$$\Sigma_D = 0.434C \int_{r_1}^{r_2} \frac{\sigma_D(r)\, dr}{r^{\nu+1}} \tag{8}$$

The radar cross section has been numerically integrated for different values of the exponent ν and lower radius limit r_1. The upper limit $a = 30$ is fixed and corresponds to $r_2 = 3.3\ \mu$ at ruby wavelengths. The results for $\nu = 3.5$, $r = 0.275\ \mu$, and $r_2 = 3.3\ \mu$ with $\nu = 0.6943\ \mu$ are shown in Fig. 2. The diagram shows N_D, the total number of particles necessary with $r_1 \leq r \leq r_2$ for the cross section of the aerosols to equal the cross section of molecules at the indicated altitudes. While other results will be presented elsewhere, we point out that the backscattering intensity functions decrease very rapidly with decreasing particle

size, and the smaller particles do not make a significant contribution to the backscattered radiation unless they are present in very large amounts; thus a detailed description of the size distribution in the size range smaller than r_1 is unnecessary for the numerical integrations when r_1 represents the radius at which a "break" in the observed exponential distribution function is found. Also, calculations for the refractive index $\tilde{n} = 1.33$, appropriate to water droplets, gave estimates of particle concentration exceeding those obtained for $\tilde{n} = 1.5$ by a factor of approximately 3. This indicates that the use of Mie calculations for a 1.33 refractive index to interpret optical radar returns from the stratosphere (Deirmendjian, 1965) may overestimate the particle concentration by this amount if particles are indeed composed of the sulfates that have been detected in the impactor samples.

With the laser systems used in this study, the energy transmitted, W_t, was not constant from pulse to pulse; although W_t could in principle be determined by an appropriate monitoring system, this was done only occasionally. Similarly, k_t and k_r, the efficiencies of the transmitting and receiving systems, respectively, could also be measured and utilized in the calculation. In general, the atmospheric transmission coefficient, k_a, is a function of hight and time.

In the attenuation model proposed by Elterman (1964) atmospheric optical parameters have been calculated that include attenuation by molecular and aerosol scattering and by ozone absorption. The parameters computed for 0.7-μ radiation indicate that the k_a^2 term would not vary by more than 3% over the entire altitude interval from 10 km to 30 km. In reality, much larger values of the attenuation could be caused by the presence of thin tropospheric clouds that might not be easily detected visually. While the optical radar is, of course, capable of detecting such stratifications, the important point is that the beam suffers almost all of its attenuation in the troposhere; k_a is taken to be, for our purposes, a

function of time but not a function of height.

The results of one of the earlier observations have been published previously (Ficocco and Grams, 1964) and are presented in Fig. 3 with a change in the symbols used to describe the scattering ratio. Curve a is the observed optical radar return obtained by averaging 20 consecutive traces. The expected return from a dust-free atmosphere is plotted as curve b. The scattering ratio Σ/Σ_M is curve c, the ratio between these two curves. Curve b has been shifted in the vertical direction to agree with the experimental curve a at the extreme ranges.

The scattering ratio shows a maximum near 20 km in agreement with the results of Junge et al., and the broadscale features of the profile are in agreement with profiles derived by other optical techniques (Volz and Goody, 1962; Bigg, 1964; Elterman and Campbell, 1964). From the scattering computations reported above we infer that at the altitude of the maximum scattering ratio the equivalent particle concentration of spheres with a refractive index $n = 1.5$ and $0.275\,\mu \leq r \leq 3.3\,\mu$ is $\sim 1\ cm^{-3}$. This estimated concentration is an order of magnitude larger than the results of the particle collections of Junge and his collaborators, although it agrees with the photoelectric particle counts obtained by Rosen (1964). The large increase in particle concentration can be attributed to the perturbed conditions existing after the eruption of the Mount Agung, which we shall discuss below. In the reduction of the present data we have determined the constant terms of Eq. (1) by using observed optical radar returns to calibrate the apparatus. The vertical coordinates of a number of consecutive traces are averaged to eliminate statistical fluctuations of the signal. Ratios are then computed between the average optical radar return and the values calculated from the optical radar equation for an arbitrary radar constant and for a model, dust-free atmosphere by using tabulated densities in the U.S. Standard Atmosphere, 1962. Then these ratios are normalized by dividing each ratio by the average calculated ratio for the altitudes between 25 km and 30 km. The resulting numbers are taken to be the scattering ratio, called Σ/Σ_M and interpreted as the ratio of observed to molecular cross sections. The

normalizing procedure assumes that the average value of the scattering ratio for altitudes between 25 km and 30 km is unity.

This assumption, although supported by the results of Rosenberg, Sandomirsky, and Poldmaa (1966), introduces the possibility that the deduced scattering ratios may be larger, that is, the technique may underestimate the dust content of the stratosphere. The inherent noisiness of the signal, especially in the case of the weaker echoes from higher altitudes, could lead to anomolously large scattering ratios throughout the entire calibration interval; this may result in scattering ratios less than unity below 25 km if appropriate modifications are not incorporated into the calibration procedure. The computation scheme, therefore, tested the scattering ratios for the region below 25 km; if any value was found to be less than one, it was utilized to recalculate the scattering ratio profile to obtain a minimum of unity below 25 km.

In some of the early 1964 data it was necessary to incorporate a correction to the echoes observed from the lowest altitudes to account for the slow opening time of the receiver shutter. The correction causes the adjusted scattering ratios to tend toward the average values for the study period. The correction was applied to a limited number of profiles and to the low-altitude part of the profile only. Results affected by the correction are indicated in the text.

Another potential source of error lies in the use of a model molecular atmosphere for computing the expected optical radar echo. A cursory examination of temperature profiles for Bedford, Massachusetts (plotted, for example, on the ozonagrams of Hering and Borden, 1965) indicates that density fluctuations at altitudes between 10 km and 30 km seldom exceed 5% of the molecular density specified in the U.S. Standard Atmosphere, 1962. Variations in atmospheric density were therefore neglected to maintain the logistic simplicity of the observations, and are recognized as a source of uncertainty.

5. CHARACTER OF THE RETURNS

Optical radar observations of stratospheric aerosols were made from January 1964 to August 1965 on as many nights as acceptable viewing conditions and the existence of properly functioning instrumentation permitted. Most of these observations were conducted at Lexington, Massachusetts (42°25'N, 71°15'W); some data were also obtained at College, Alaska (64°53'N, 143°3'W), simultaneously with studies of noctilucent clouds during the summer of 1964 (Fiocco and Grams, 1966). The reduced data cover 66 nights of observation in Massachusetts and 11 nights in Alaska. Only a fraction of the available records has been digitized and analyzed; we have utilized almost 7000 of approximately 50,000 traces obtained in Massachusetts, although most of the data from Alaska have been reduced.

Since the primary purpose of the study is to evaluate the average and time-variant characteristics of the dust layer, it is necessary to separate the fluctuations attributable to the variable properties of the dust layer from those that can be ascribed to the method of detection. The shape of the observed traces exhibits a great deal of variability from trace to trace; this variation has the appearance of random fluctuations superimposed on a mean return. As an example, more than 200 consecutive traces, taken at 5-second intervals on 18 December 1964 were digitized and used to study these fluctuations. The mean and standard deviations of the digitized coordinates at each altitude were calculated. If the variations in the photoelectron flux recorded on the oscilloscope traces are indeed random variations, the calculated values of the standard deviation should vary as $\sqrt{\frac{dn}{dR}}$, where $\frac{dn}{dR}$ is the observed photoelectron flux at the photocathode per unit interval of height. The computed values of the mean and standard deviations of the optical radar return at each altitude are plotted in Fig. 4 on logarithmic graph paper. The points lie reasonably close to a line with the slope equal to 1/2. Other calculations also corroborate a conclusion that for that particular set of observations the fluctuations are random. For example, autocorrelation coefficients at one lag, that is, computed for delays of 5 seconds,

were calculated at each altitude and found to be very small, with a mean value of almost zero and having absolute values $\lesssim 0.1$. These uncorrelated fluctuations are believed to be primarily due to shot noise affecting the photomultiplier current, and also, possibly, to phase effects in the propagation of the wave; it is considered unlikely that they represent variations in scattering particle concentrations.

Uncertainties associated with statistical fluctuations of the signal can be eliminated by integrating a sufficiently large number of optical radar returns. The effect of this in reducing the random fluctuations of the signal is summarized in Fig. 5, which shows isopleths of rms fluctuations of the echoes received from 0.5-km layers as a function of the number of traces added together and of the altitude of the echo. The fluctuations are expressed as percentage deviations from the mean photoelectron flux observed at each altitude. The curves represent smoothed values of the results of computations performed on the same series of digitized traces that were used to construct Fig. 4. The expected rms fluctuations do, of course, increase with altitude and with smaller numbers of traces used to compute the mean. This figure indicates the expected error associated with the experimental returns for 0.5-km altitude intervals. If, indeed, the fluctuations are random and if, also, they are not correlated with fluctuations at contiguous altitudes, values appropriate to 1-km averages should be reduced by a factor $\frac{1}{\sqrt{2}}$. The values have been calculated for a single night of observation only. They can be used as an indication of the statistical errors in the experimental results for the entire series, although day-to-day variations in the intensity of the echoes change the expected rms fluctuations for each altitude (see Fig. 4).

Some typical results of the dust study are presented in graphical form in Figs. 6, 7, and 8. Fig. 6 shows 5 dust profiles obtained during a 15-minute interval on 18-19 December 1964. Each profile is constructed by averaging 35 optical radar traces recorded at 5-second intervals; the profiles were therefore obtained every 3 minutes. Since the constants for the radar equation are subject to some uncertainty, the profiles have been constructed with the scattering ratio $\frac{\Sigma}{\Sigma_M}$ plotted on a logarithmic

scale. The semi-logarithmic presentation provides an invariant shape for the profile, since change in the radar constant will only shift the curve in the horizontal direction. The enhanced backscattering from the 15-20 km region is quite evident, showing little qualitative change from one profile to the next. The appreciable differences between consecutive profiles at higher altitudes are attributed to the statistical fluctuations of the signal and are not used to derive a wave structure for thin aerosol layers at these altitudes, as suggested by Collis and Ligda (1966) with similar apparatus. We point out that the intensity of radar echoes at 30 km is approximately two orders of magnitude less than those observed at 10 km, and the observed profiles at the higher altitudes are therefore more sensitive to the shot noise of the photodetector and other sources of statistical fluctuation (see Fig. 5).

Fig. 7 presents 10 hourly profiles of the scattering ratio for a single night of observation on 11-12 March 1965. These profiles are constructed by averaging 25 consecutive traces recorded at 2-second intervals; each series, therefore, was obtained in slightly less than one minute. These profiles and those from other nights of observation also show little qualitative change in the 15-20 km region, even on an hourly scale, thereby suggesting that the aerosol layer is a slowly changing feature of the lower stratosphere. Again, returns from higher altitudes are subject to a considerable amount of statistical fluctuation. Fig. 8 shows daily profiles obtained on 5 consecutive days, in March 1965. The daily profiles are produced by averaging all hourly profiles obtained during each night of observation. These do indicate some changing features of the vertical distribution of dust during 5 days.

The average vertical distribution of dust for all analyzed data obtained during 1964 and 1965 is illustrated by the solid line in Fig. 9. A maximum value for the scattering ratio $\frac{\Sigma}{\Sigma_M} = 1.9$ is obtained. The dotted lines indicate the extent of the rms deviation of the daily profile. Such deviations are essentially due to the varying characteristics of the layer; and in much smaller part to the statistical fluctuations of the measuring technique. We

estimate that the magnitude of such instrumental fluctuations varies from 0.2% to 3% of the observed scattering ratios throughout the altitude range of interest, and, in any case, we regard them as negligible with respect to the intrinsic fluctuations of the stratospheric dust. We must, however, point out that small systematic errors may still affect the measurements, especially in regard to the assumption that the 25-30 km region is adequately described by our model dust-free atmosphere. The presence of appreciable amounts of dust in this region would cause us to infer dust amounts smaller than the actual dust content of the stratosphere.

To illustrate the seasonal changes that were observed in the dust layer during the study, bimonthly means for all of the daily profiles have been calculated by grouping the appropriate daily profiles. The results are presented in Fig. 10, which shows isopleths of the scattering ratio $\frac{\Sigma}{\Sigma_M}$ as a function of altitude and time. Fig. 11 indicates day-to-day fluctuations of dust amounts at 16 km, near the center of mass of the aerosol layer. Points plotted as open circles on this diagram and all subsequent diagrams refer to dust observations that incorporate an instrumental correction. The data collection was interrupted from mid-February 1964 until April 1964. Another break in the collection of data at Lexington occurred in the summer of 1964 when observations of noctilucent clouds were performed; data for the summertime high-latitude stratosphere were, however, obtained during that period. The observed dust amounts exhibit an appreciable amount of scatter with a generally decreasing trend. The high initial aerosol concentration, followed by decreased amounts during the spring of 1964, follows the same trend as the twilight observations of Volz (1965) during the same period. Fall 1964 was also a period of high aerosol concentration, with decreasing dust amounts indicated during early winter 1964/65. Except for a short period of low dust amounts during mid-March 1965, the aerosol concentration increased again during the spring of 1965. A subsequent decrease during the summer of 1965 suggests that the stratospheric aerosol layer may be returning to normal conditions. Low dust amounts during the summer of 1965 have also been found by Volz (1965) and are corroborated by other investigations such as studies of the small-ion density

in the stratosphere (Paltridge, 1966). The considerable length of time required for the turbidity of the stratosphere to return to normal conditions is comparable to the well-documented Krakatoa eruption in 1883 (Wexler, 1951).

Rosen (1964) compared the particle concentrations at the concentration maximum of the aerosol layer with particle concentrations at 85 km. He concludes that the source must lie at least above 85 km, and that the particles are probably of extraterrestrial origin. In view of the composition of the particles (Friend et al., 1961; Junge et al., 1961) and the omnipresent minimum scattering ratio above 20 km observed in this study, however, it appears that the particles may be formed within the layer as suggested by Junge et al. or, at least, can be associated with the spatial distribution of the debris injected into the stratosphere by the eruption of the Agung volcano.

Various statistical calculations have been performed with the dust data. Using daily values of the scattering ratios averaged over 1-km altitude intervals, we calculated autocorrelation coefficients at each altitude for lags up to 30 days; with the 66 different profiles available for the statistical calculations approximately 15 data pairs could be assembled for each lag category. The curves, shown in Fig. 13a for dust measurements at 14, 16, 18, and 20 km, indicate large values of the autocorrelation coefficients for lags up to 6 days. Unfortunately, the autocorrelation function was calculated from an inhomogeneous sample in which the number of data pairs decreases as the time lag increases, so that the statistical reliability of the correlations decreases for the larger lags. Fig. 12b indicates the number of pairs utilized for each value of the lag parameter.

The profiles obtained during the summer of 1964 in Alaska are shown in Fig. 13. The average profile for the summer is also included in the diagram. An average scattering ratio of ~1.7 with daily rms fluctuations ~0.15 is observed at the altitude of the maximum scattering ratio. The amounts were smaller and located at slightly lower altitudes than those obtained in Massachusetts. Other observations (Manson and Junge, 1961;

during the time period from January 1964 to February 1965. Fig. 16 is a scatter diagram with the center of mass of the aerosol layer as the ordinate and the corresponding tropopause height at the radiosonde station as the abscissa. No pronounced trend is evident, although the regression line drawn through the points on the scatter diagram has a slope of approximately +0.1, thereby indicating that changes in the tropopause height can have a small effect on the distribution of mass in the aerosol layer. Previous observations by Manson and Junge (1961), also do not exhibit pronounced changes in the vertical distribution of aerosols following daily changes in the height of the tropopause.

6. CORRELATIONS WITH OZONE

Daily fluctuations of the stratospheric dust content have been compared with those of atmospheric ozone. Most ozone variations occur in the lower stratosphere, where maximum concentrations are observed. Ozone is generally considered to be a photochemically quasi-conservative quantity in the lower stratosphere and has frequently been used as a tracer to indicate atmospheric motions. Various aspects of ozone research have been reviewed by Craig (1965) and by Vassey (1965), among other authors.

In the usual method for determining the total amount of ozone in a vertical column a comparison is made of the flux of solar radiation at different wavelengths in a spectral region where solar energy is absorbed by ozone but still reaches the surface of the earth with sufficient intensity to be detected by a spectrophotometer (Dobson, 1957). The temporal and spatial distribution of total ozone has been studied for several decades, and although daily fluctuations of total ozone often exceed seasonal changes, the climatological data obtained by averaging large numbers of observations show a maximum amount in spring and a minimum in fall, with the largest seasonal fluctuations observed at high latitudes. Many methods of observing the vertical distribution of ozone are also available (Craig, 1965). Systematic observations of the vertical ozone distributions have been performed at a network of North American Stations, since January 1963, in a program organized by the Air Force Cambridge Research Laboratories (Hering, 1964; Hering and Borden, 1964, 1965a). A chemiluminescent device designed by Regener (1964) has been used in the ozonesonde network. The balloon-borne instrumentation provides detailed profiles of the vertical distribution of ozone; absolute calibration of the apparatus relies on a surface measurement of the total ozone with the Dobson spectrophotometer. An analysis of the reliability of the ozonesonde measurements has been presented by Hering and Dutch (1965).

Ozone and dust amounts have been correlated with dust profiles derived from the optical radar observations and ozone data supplied by

W. Hering and T. Borden. . These ozone data included vertical ozone profiles obtained at Bedford, Massachusetts, as part of the regular ozonesonde program, as well as more numerous complementary data on the total ozone amounts measured daily at Bedford.

Dust and ozone profiles have been expressed in terms of their average concentration in 1-km altitude intervals in the lower stratosphere, and correlation coefficients were then calculated for each altitude interval. The results are given in Fig. 17 which shows the correlation between dust and ozone for altitudes from 12 km to 24 km for several different time lags between respective measurements. The ozone profiles had been obtained according to a schedule providing weekly observations and some additional observations in situations of special meteorological interest. The observation dates for dust and ozone did not always coincide, since attempts to correlate the two parameters were not initiated until the fall of 1964. Furthermore, the optical radar observations are dependent on visibility conditions and could not always be performed at times near the scheduled ozone soundings. Nevertheless, approximately 15-20 pairs of ozone and dust profiles could be assembled for various time lags by using the 66 dust observations and the available ozone profiles. Simultaneous data did not exist; optical radar observations were performed at night, typically near 2100 EST, while the ozone observations were usually scheduled at 0700EST. Thus, the observations were normally obtained approximately half a day apart. The correlation coefficients displayed in Fig. 17 are crosscorrelations at each altitude between ozone and dust with dust measurements approximately 0.5 and 1.5 days before and after the ozone observation. The correlation coefficients for altitudes near the center of mass of the aerosol layer are all negative, having values of approximately -0.5 for the lag categories shown in the diagram. The correlations were extended to 30-day lag in order to establish the statistical properties of presumably uncorrelated data. The value -0.5, which was obtained for the coefficient at altitudes near the center of mass of the aerosol layer,

was thereby found to be ~1.9 standard deviations from the mean value of the correlation coefficient for large lags. Since the probability of exceeding 1.9 standard deviations is approximately 6% for a normal distribution, we believe that the negative value of the correlation coefficient is physically significant.

The aerosol data have also been correlated with the measurements of total ozone obtained at Bedford, Massachusetts, to provide additional statistical evidence of a relation between the aerosol layer and stratospheric ozone. Most changes in total ozone are highly correlated with changes in the ozone content of the lower stratosphere. Mateer and Godson (1960) found, for example, a correlation coefficient of +0.97 between total ozone and the ozone in the 12-24 km altitude interval for a Canadian station. Hering and Borden (1965b) have correlated ozone concentrations obtained from the ozonesonde network with total ozone; the highest correlation coefficients were usually obtained at altitudes between 8 km and 20 km.

Since total ozone observations are performed on a daily rather than on a weekly basis, a larger number of data pairs was available and the statistical significance of the results is thereby increased. The results are summarized in Fig. 18, which shows isopleths of constant correlation coefficients as a function of the altitude of the dust measurement and the lag, in days, between the dust and total ozone observations. Again, simultaneous observations were not obtained because the total ozone measurements were taken near 1200 EST. The calculations show a very high anticorrelation between total ozone and the dust at 16 km (the center of mass of the dust layer) which falls off rapidly with increasing lag. The computed correlation coefficient is -0.7, which exceeds the mean correlation coefficient at large lags by almost 4 standard deviations. This result verifies the relationship between stratospheric dust and ozone during the study period.

Fig. 19 is a scatter diagram showing observed values of $\frac{\Sigma}{\Sigma_M}$ at 16 km and total ozone for data pairs with dust measured the night after ozone; if ozone data were missing for that lag category, the ozone observation obtained the day after the dust measurement was used in the diagram. Total ozone is plotted as the ordinate and the dust parameter as the

abscissa in the display; data pairs in which the dust parameter has a shutter correction are plotted as open circles. The negative correlation is evident in the figure, with the largest dust amounts associated with low ozone values and maximum ozone amounts observed only when dust concentrations are low.

It has been pointed out, in reference to Fig. 11, that high dust amounts were observed in the fall of 1964 and some low values were obtained in the spring of 1964 and of 1965. Since the total ozone measurement always exhibits characteristically high values in spring and low values in fall, the correlation coefficients were recalculated; ozone data were used with the seasonal variation of ozone removed, to eliminate the possibility that the correlation was fortuitously obtained by the incursion of large amounts of dust from the Southern Hemisphere during seasons with a minimum ozone content and subsequent decreasing trends of dust concentration during seasons with increased ozone amounts. Thirty-day running means were subtracted from the observed total ozone amounts to eliminate seasonal variations of ozone. The correlation coefficients calculated for this case are shown in Fig. 20. As would be expected with regard to the seasonal effects that have been described, the magnitudes of the correlation coefficients were reduced; however, the dust amounts at altitudes near the center of mass of the aerosol layer and total ozone remain negatively correlated for lags up to two days. Observed correlation coefficients have values of approximately -0.35, exceeding the mean of correlation coefficients computed for longer lags by over two standard deviations, thereby remaining statistically significant at the 5% level of confidence. A scatter diagram for dust amounts at 16 km and deviations from the monthly mean of total ozone has been constructed and is shown as Fig. 21. Again, a negative trend in the observations is indicated with high dust amounts tending to be associated with negative deviations and large positive deviations occuring only when dust concentrations are low.

The calculations that have been described apply to the time period from January 1964 to April 1965, for which vertical profiles of

atmospheric ozone were available. Total ozone amounts were available, however, up to August 1965; data pairs with all available total ozone observations included were used in constructing the scatter diagrams shown in Figs. 19 and 21. The correlations between dust and total ozone measurements were recalculated, with dust data included for the extended time period that concurred essentially with the observed trend toward natural conditions in the stratospheric dust layer and was characterized by low dust amounts. For the dust-total ozone correlations, the coefficients were reduced by approximately 20% of their values when the data for the summer of 1965 were included. The calculations may indicate that the relationship was not as pronounced in the presence of smaller dust amounts; this is also visually observed in Figs. 25 and 27 by the trend for increased scatter in the data points associated with lower dust concentrations.

In order to compare the temporal behavior of ozone with that of the stratospheric aerosol, autocorrelation functions for ozone concentrations at each altitude have been calculated. The results are shown in Fig. 22. The considerable amount of persistence displayed by the dust data is lacking; the autocorrelation function decreases to insignificant values withing one or two days. Similar calculations performed with total ozone are shown in Fig. 23. The autocorrelation function falls rapidly, but then remains significantly correlated for at least a month; this is taken to reflect the seasonal trend in the total ozone. The calculations for Fig. 22 were made by using only the ozone profiles obtained within a few days of the dust profiles, and the statistical fluctuations associated with small data samples are evident. In the total ozone correlogram continuous observations were used for the entire period of observation; therefore it represents a larger and considerably more homogeneous sample from which to calculate the autocorrelation function.

To conclude, a significant relationship was found between ozone and aerosols in the lower stratosphere, which acts in such a way that daily

fluctuations of each parameter are negatively correlated. The aerosol study was conducted during a perturbed state of the atmosphere when particle concentrations were probably an order of magnitude larger than the natural population of stratospheric aerosols and the physical and chemical properties of the particulate material were likely to be different from the natural state. Perhaps, the observed correlation may reflect effects in the ozone measurement resulting from the perturbed conditions in the stratosphere; we are in no position, at present, to comment on the possibility of such systematic effects, which, if present, could affect both the measurement of total ozone and the measurement of the vertical distribution. The calibration technique for the Regener ozonesonde requires that integrated ozone amounts match the total ozone measurement. If the correlation is not caused by an instrumental error, it is interesting to speculate about the physical relationship between stratospheric dust and ozone. A possibility exists that the stratospheric aerosol, located in a region where ozone concentrations are much higher than near the surface of the earth, may be an important sink for the chemical decomposition of ozone through catalytic reactions (Kroening, 1965; Pittock, 1965). The two constituents may have only a passive relationship resulting from large scale quasi-horizontal mixing processes that transport the dust and ozone from different source regions. This has also been suggested recently by Pittock (1966). The various possibilities that have been discussed will be investigated in the future.

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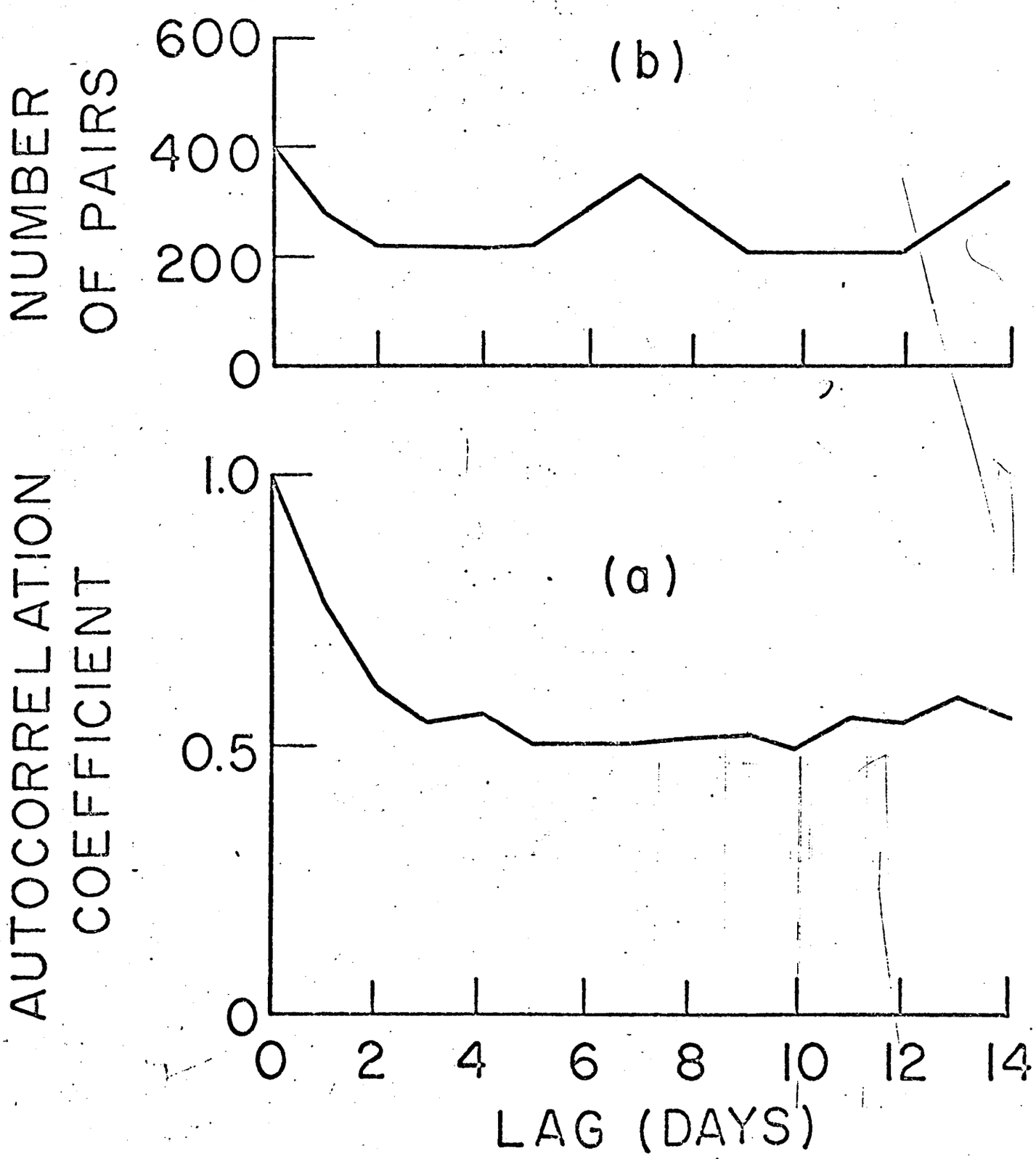

Fig. 23. (a) Autocorrelation coefficients for the total amount of ozone in a vertical atmospheric column. (b) Number of data pairs used to compute the coefficients.

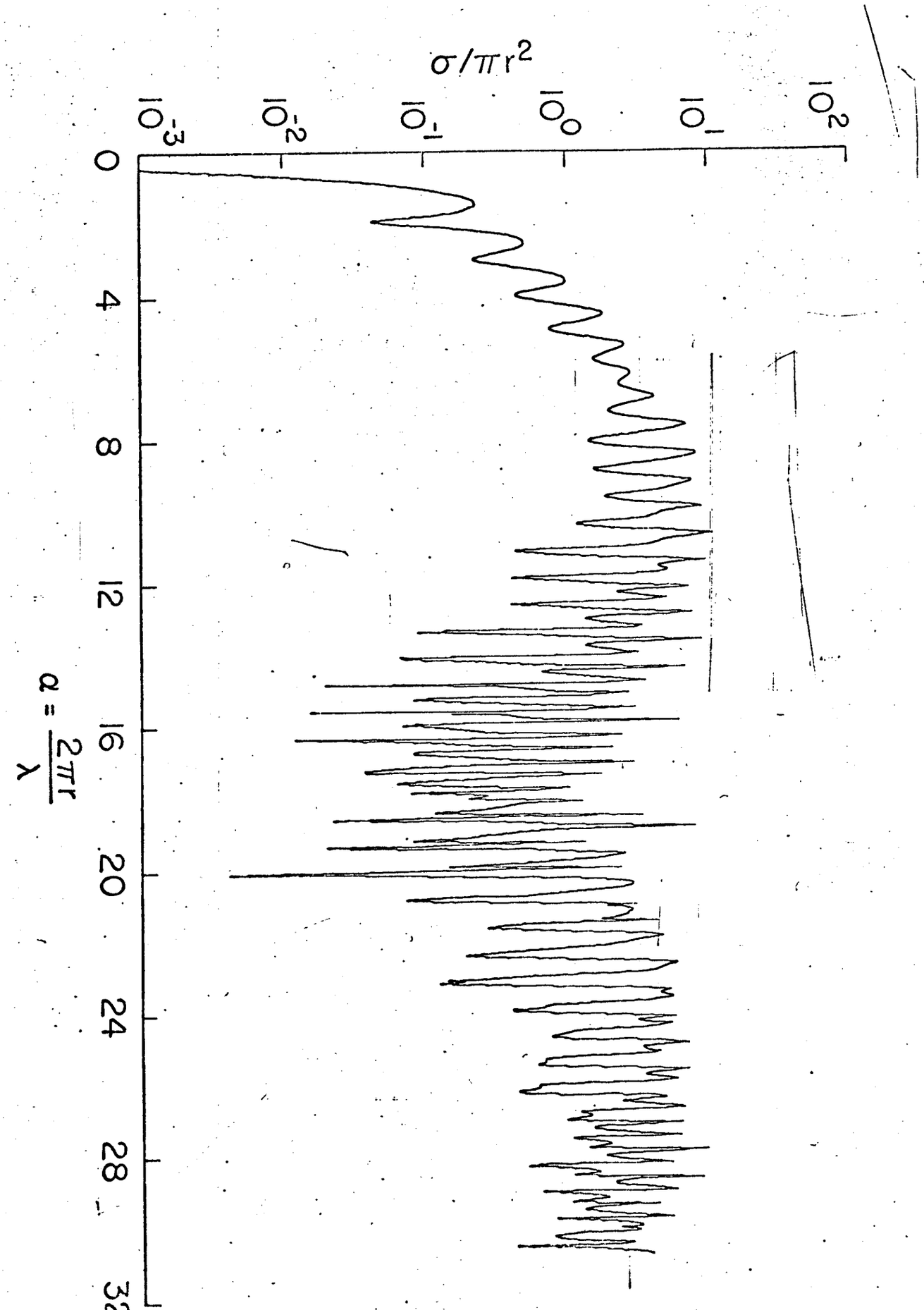

Fig. 1. Normalized radar cross section $\sigma/\pi r^2$ for spherical particles with refractive index n = 1.5.

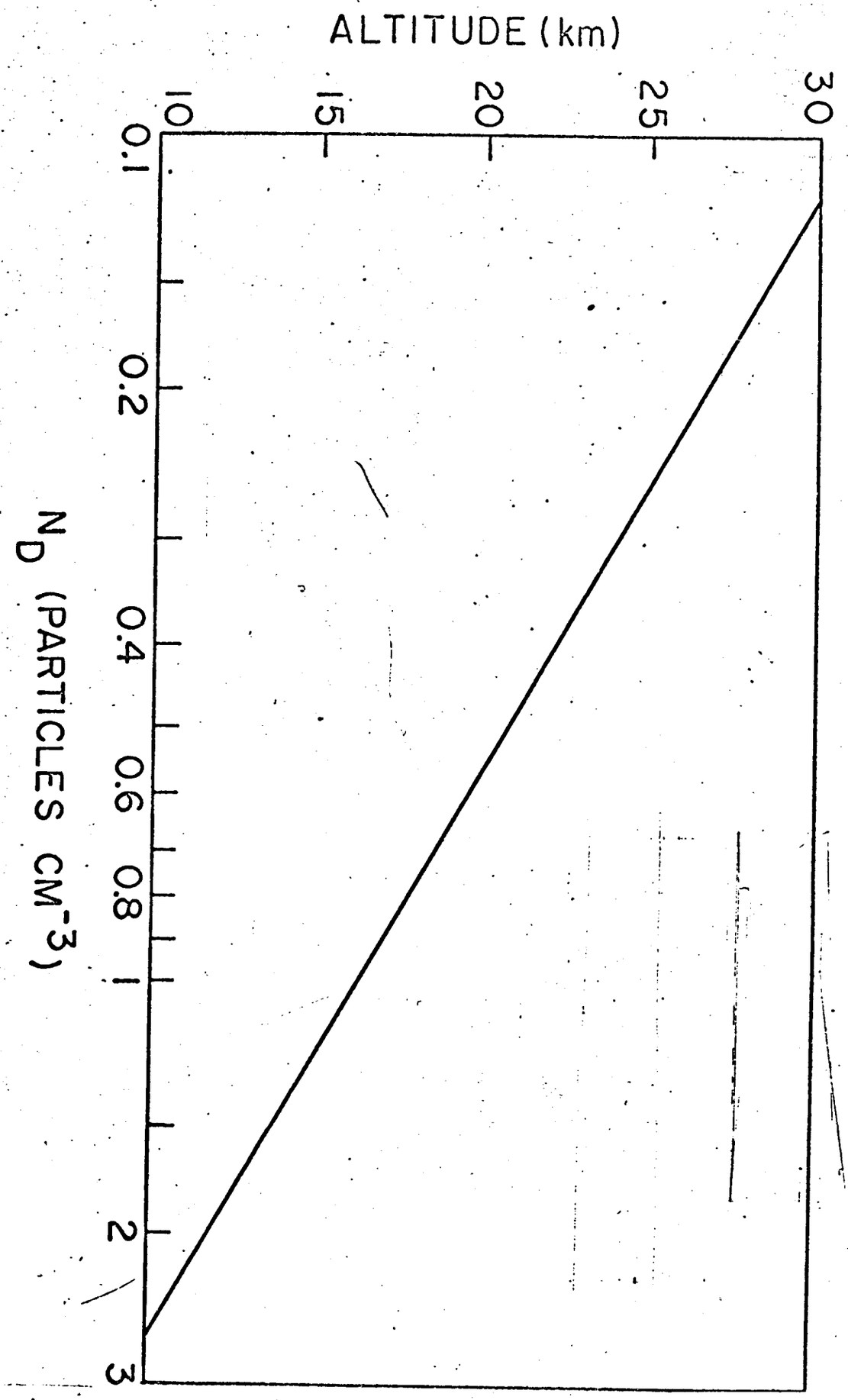

Fig. 2. Number of spherical particles with refractive index $\tilde{n} = 1.5$ required to backscatter the same amount of light as the molecular return from the lower stratosphere (computations refer to wavelength $\lambda = 0.694\ \mu$ and particle sizes specified by Eq. 6 with $\nu = 3.5$, $r_* = 0.275\ \mu$, and $r_c = 3.3\ \mu$).

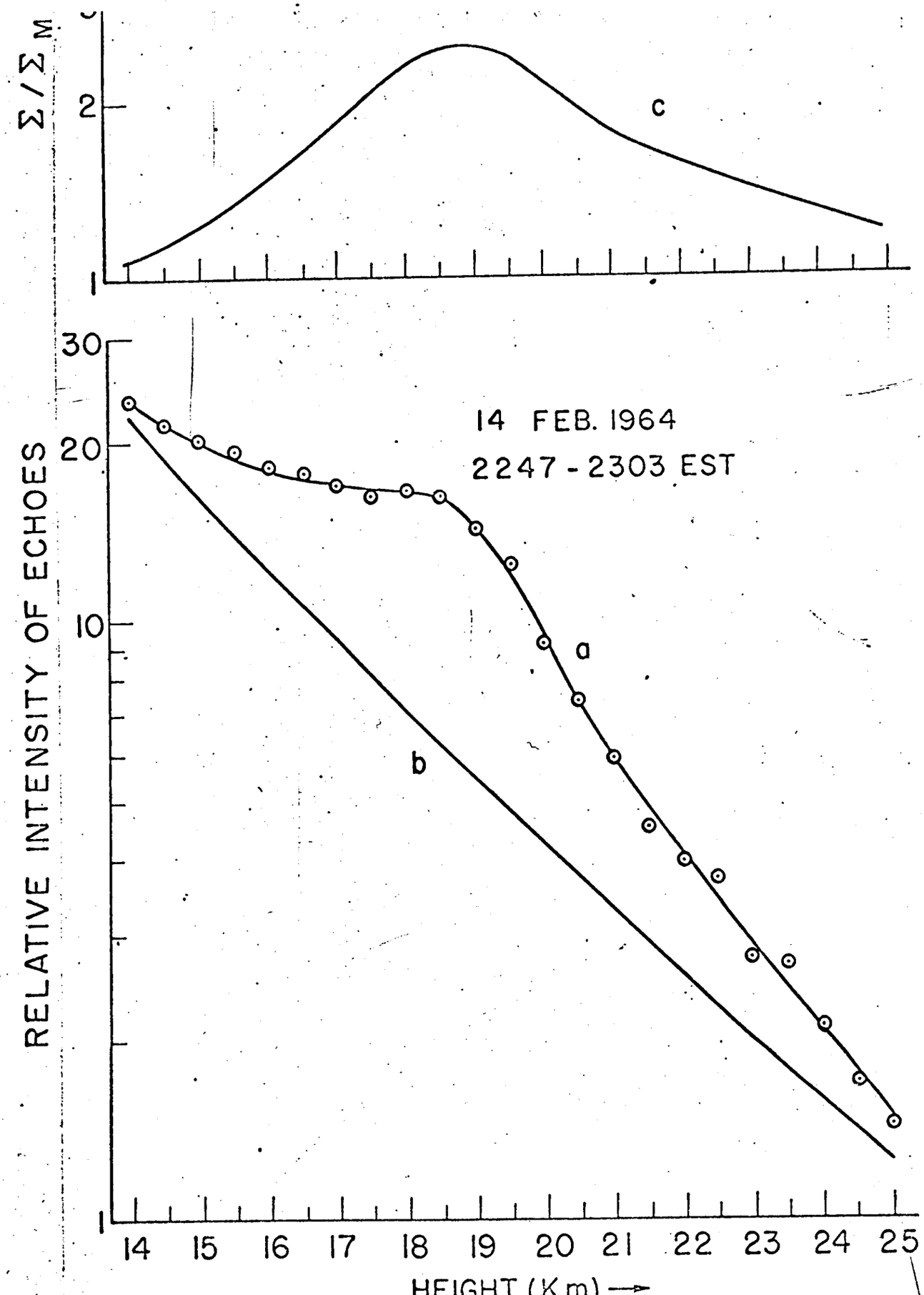

Fig. 3. Optical radar observations for 14 February 1964. Curve (a): intensity of echos from a vertically pointing optical radar. Curve (b): expected intensity of echos from a purely molecular atmosphere. Curve (c): ratio of curve (a) to curve (b), interpreted as the ratio of observed to molecular cross sections.

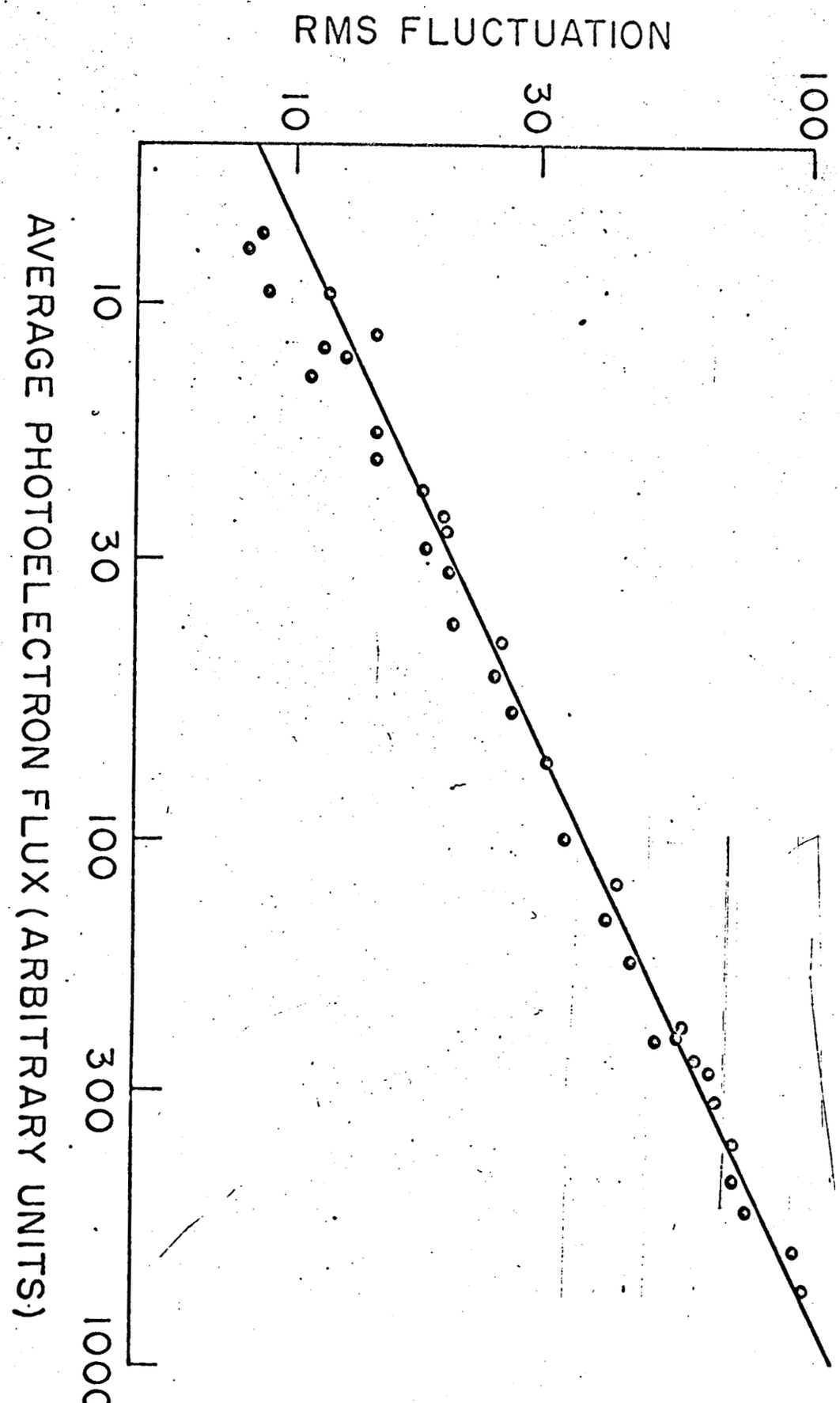

Fig. 4. Root-mean-square fluctuations of the photoelectron flux as a function of the average photoelectron flux observed for various 0.5-km altitude intervals.

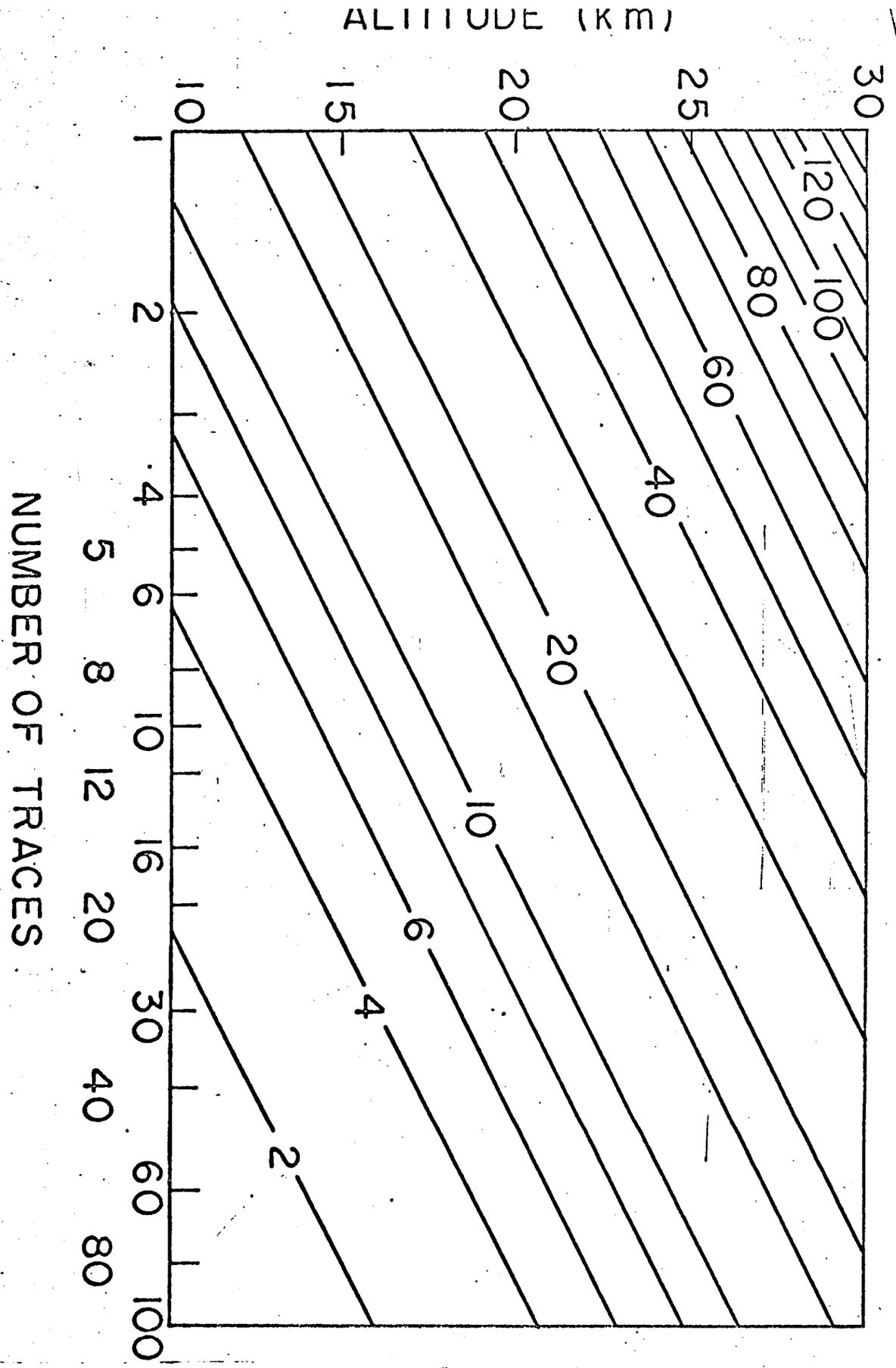

Fig. 5. Estimated rms fluctuations of the average optical radar return for 0.5-km altitude intervals in the lower stratosphere as a function of the number of individual traces used to determine the average return. The fluctuations are expressed as a percentage of the average signal for each altitude interval.

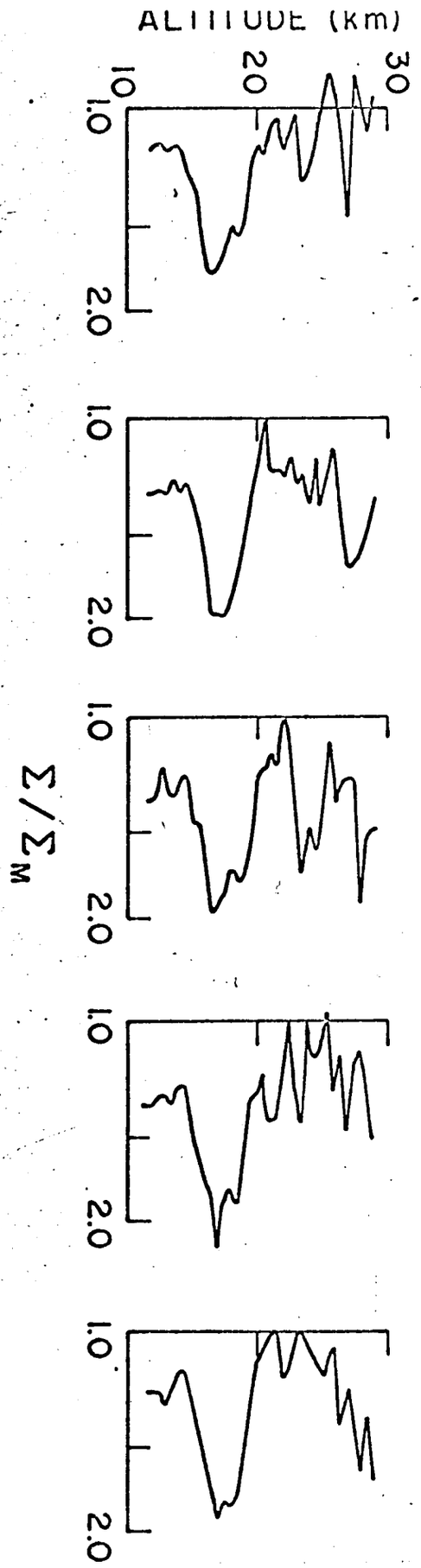

Fig. 6. Consecutive profiles obtained on 18-19 December 1964, during the time interval 2300-2315 EST. Each profile is constructed from 35 consecutive optical radar traces recorded at ~5-sec intervals.

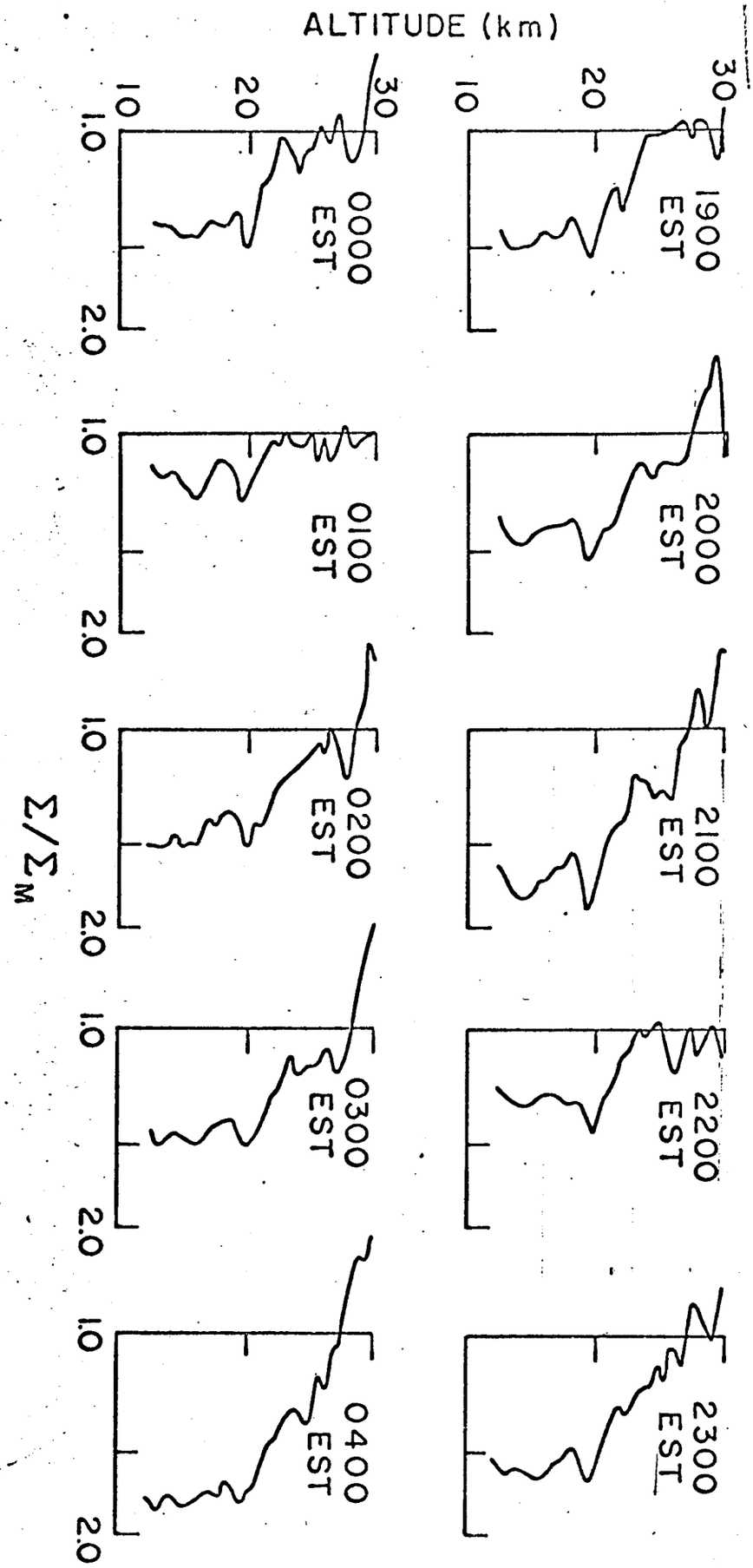

Fig. 7. Hourly profiles obtained on 11-12 March 1965. Each profile is constructed from 25 consecutive optical radar traces recorded at ~2 sec intervals.

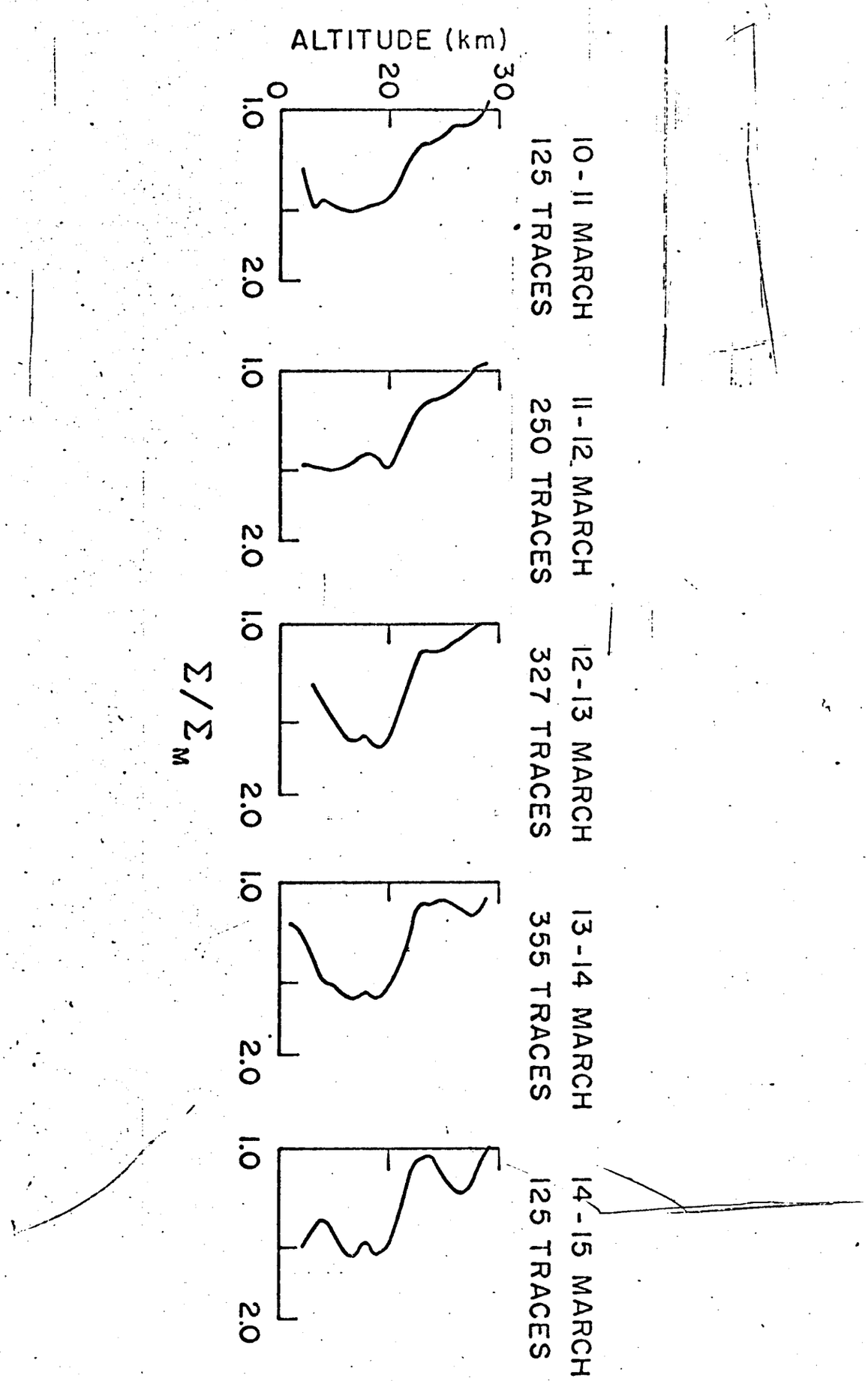

Fig. 8. Daily profiles obtained on 5 consecutive nights in March 1965.

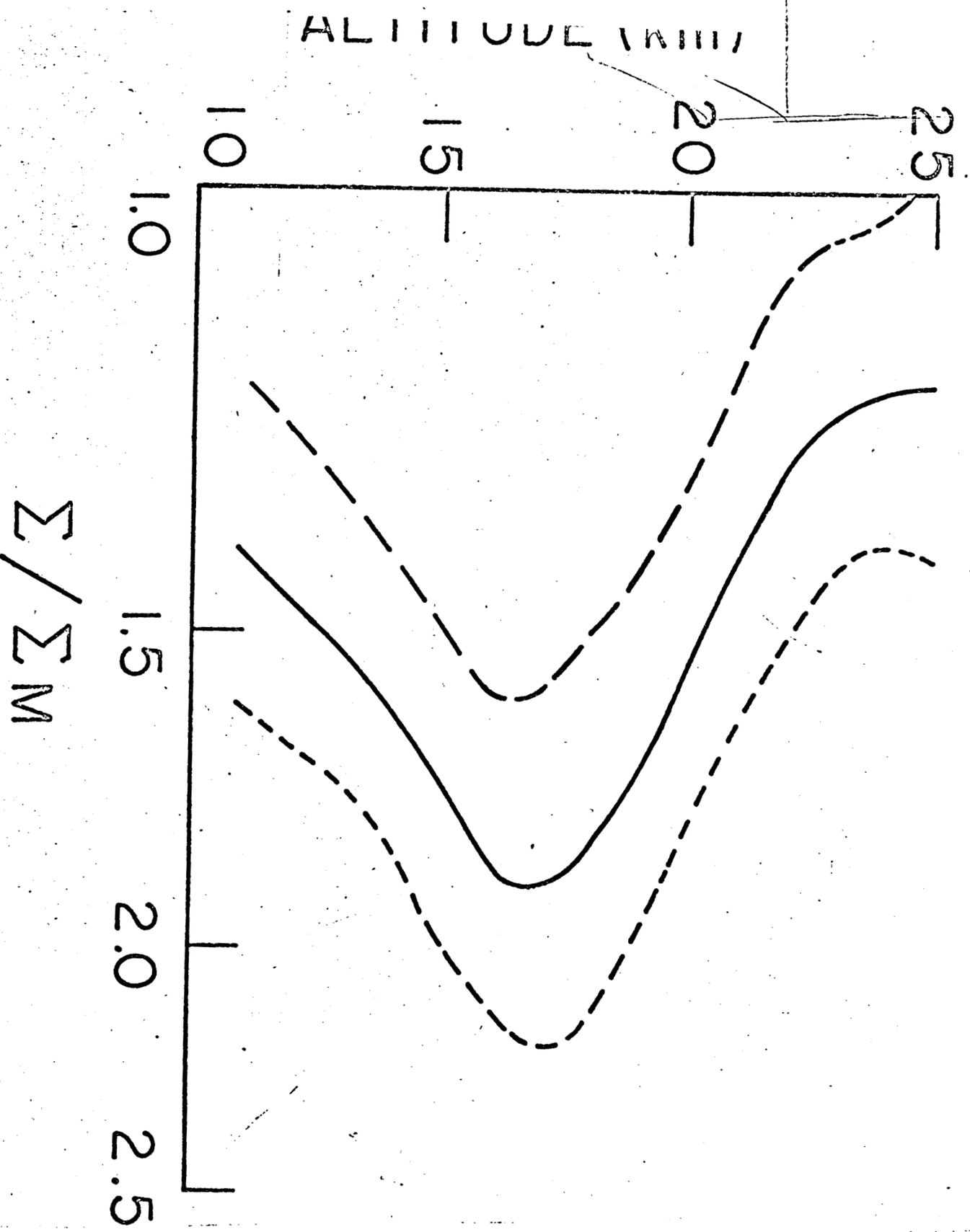

Fig. 9. Average properties of the stratospheric aerosol layer during the period of observation. The average profile displayed as a solid line, was obtained by obtaining the arithmetic mean of the 66 daily profiles used for the study; the rms deviation of the individual profiles from the mean profile is indicated by dashed lines.

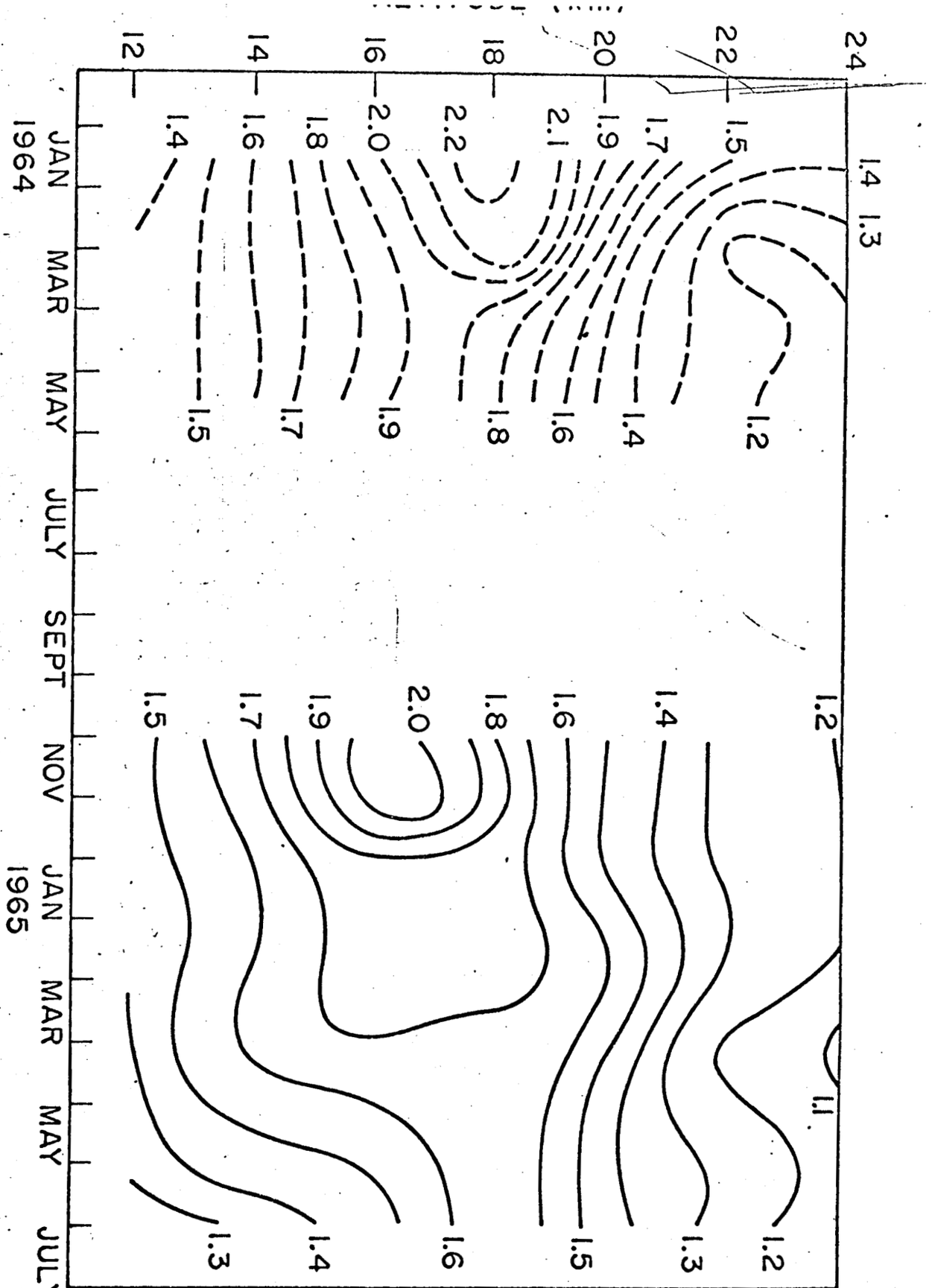

Fig. 10. Temporal variation of the stratospheric aerosol layer expressed as isopleths of mean bimonthly scattering ratios Σ/Σ_M at Lexington, Massachusetts. The dotted lines in early 1964 indicate an observation period when a relatively small number of profiles were available to compute bimonthly mean profiles.

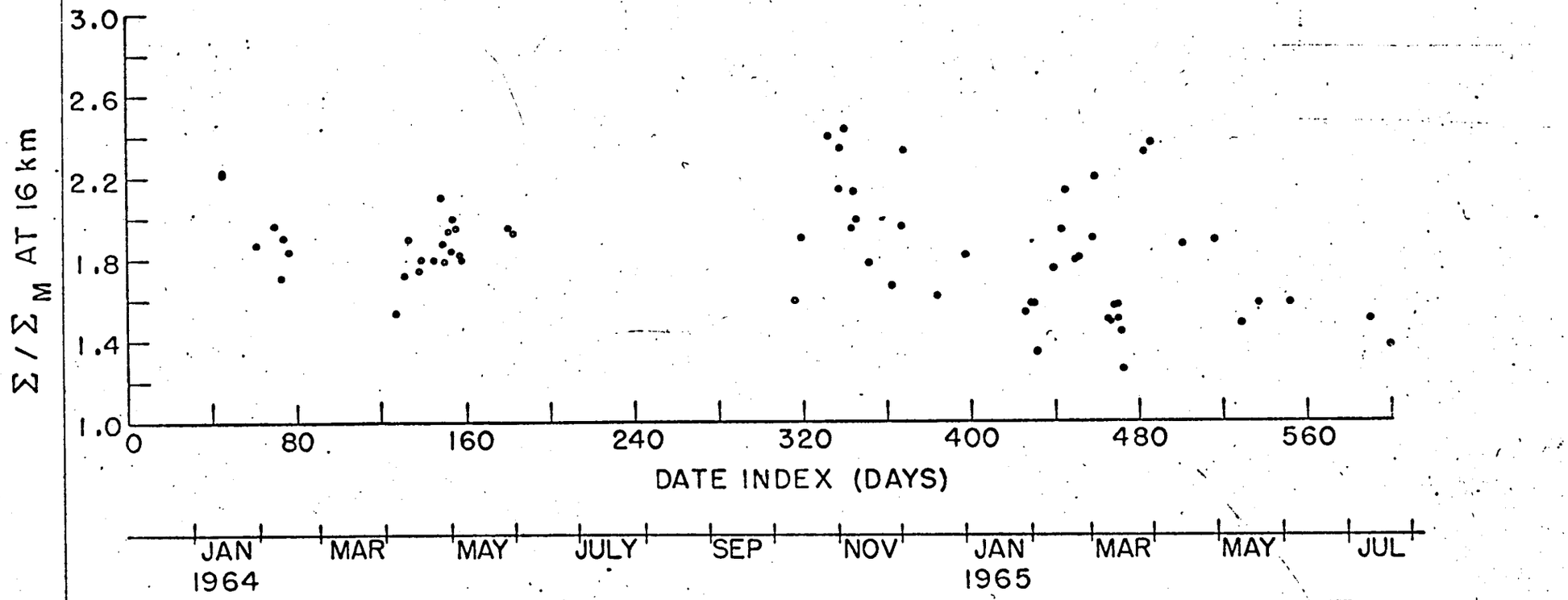

Fig. 11. Temporal variation of the stratospheric aerosol layer expressed as the day-to-day fluctuations of the scattering ratio Σ/Σ_M at 16 km at Lexington, Massachusetts. Data points plotted as open circles in this diagram and all subsequent diagrams refer to dust observations that incorporate an instrumental correction.

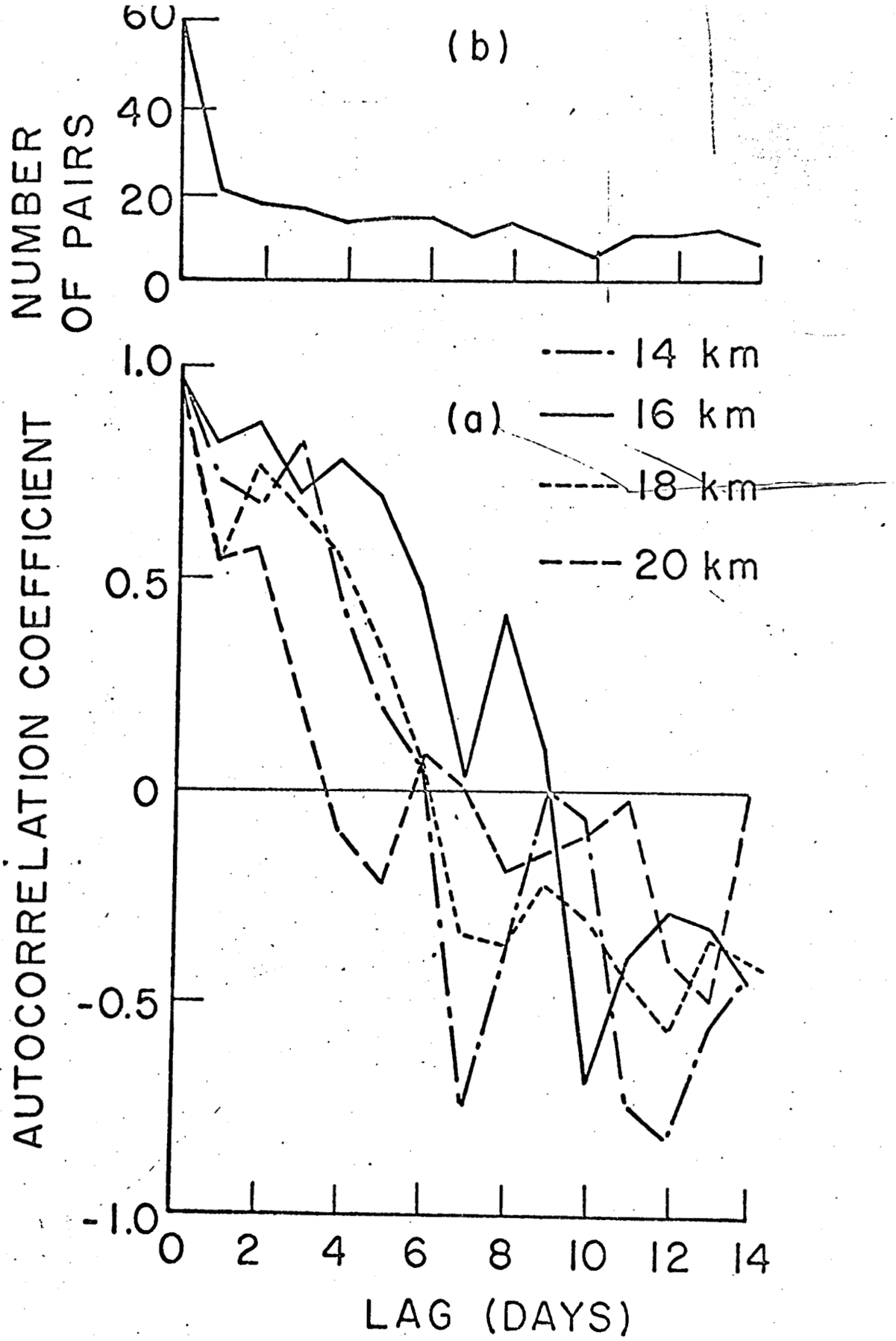

Fig. 12. (a) Autocorrelation coefficients for dust observations near the center of mass of the aerosol layer. (b) Number of data pairs used to compute the coefficients.

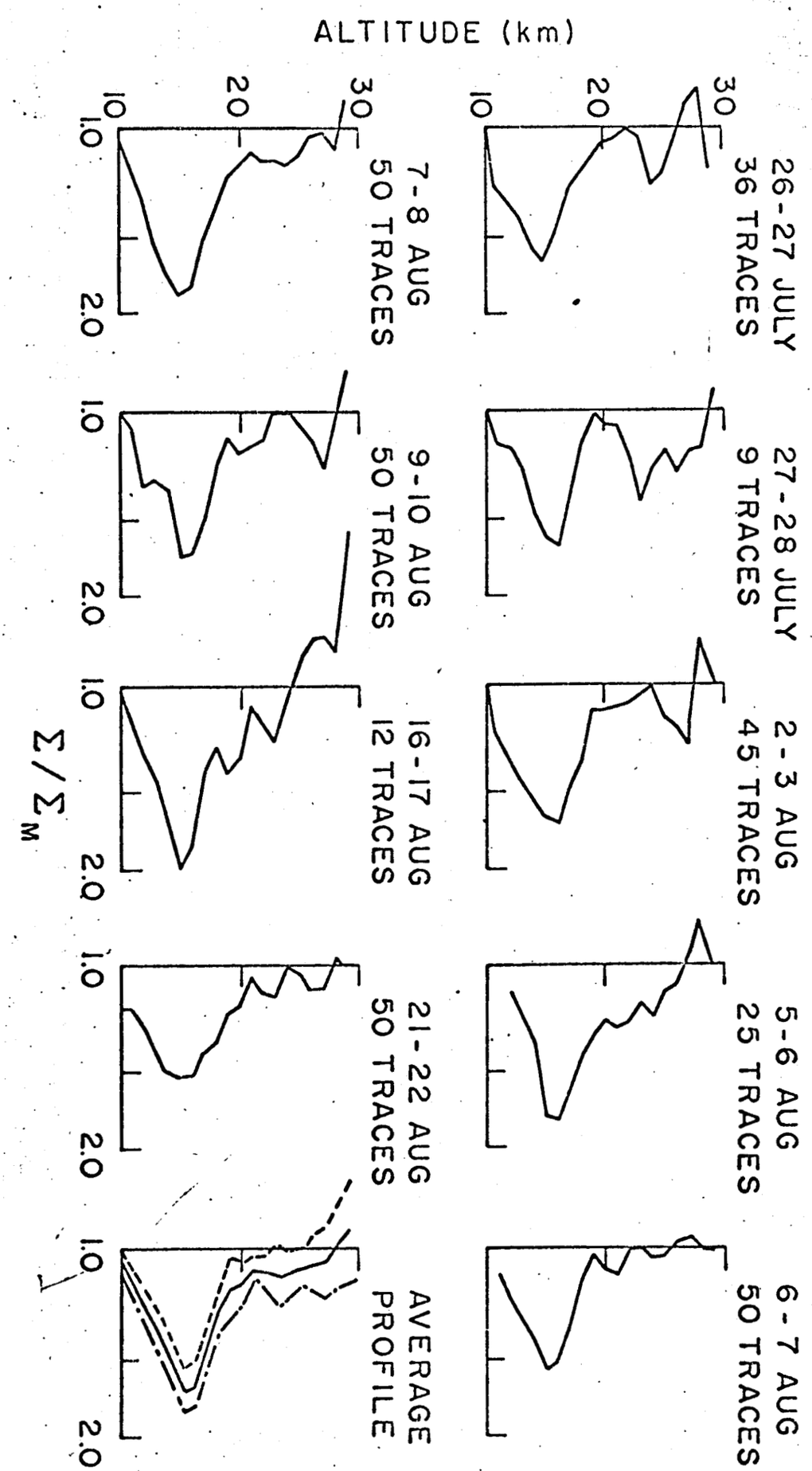

Fig. 13. Profiles obtained during summer 1964, in Alaska.

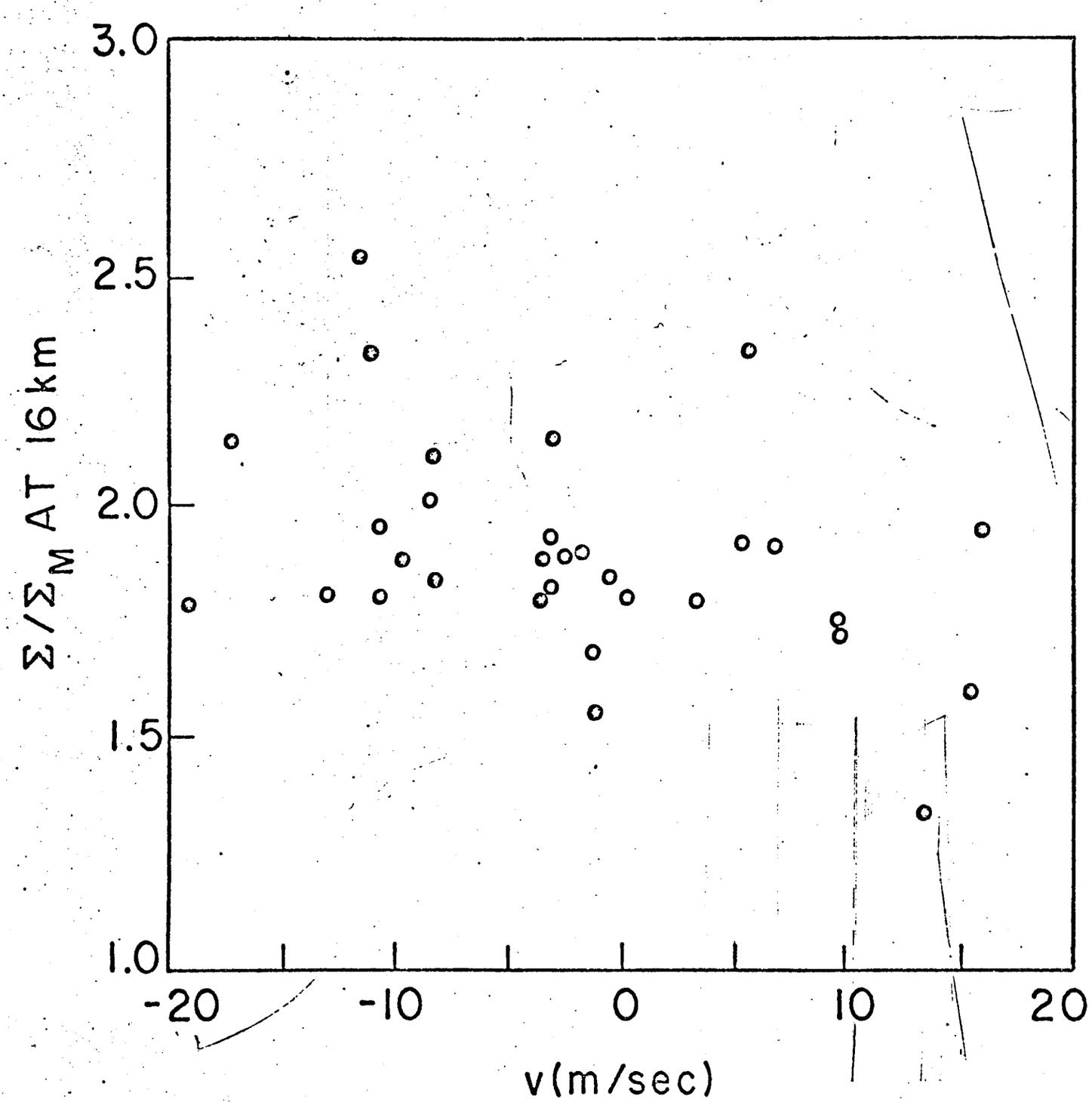

Fig. 14. Relation between the observed scattering ratio Σ/Σ_M at 16 km altitude and the northward component of wind velocity v at 100 mb.

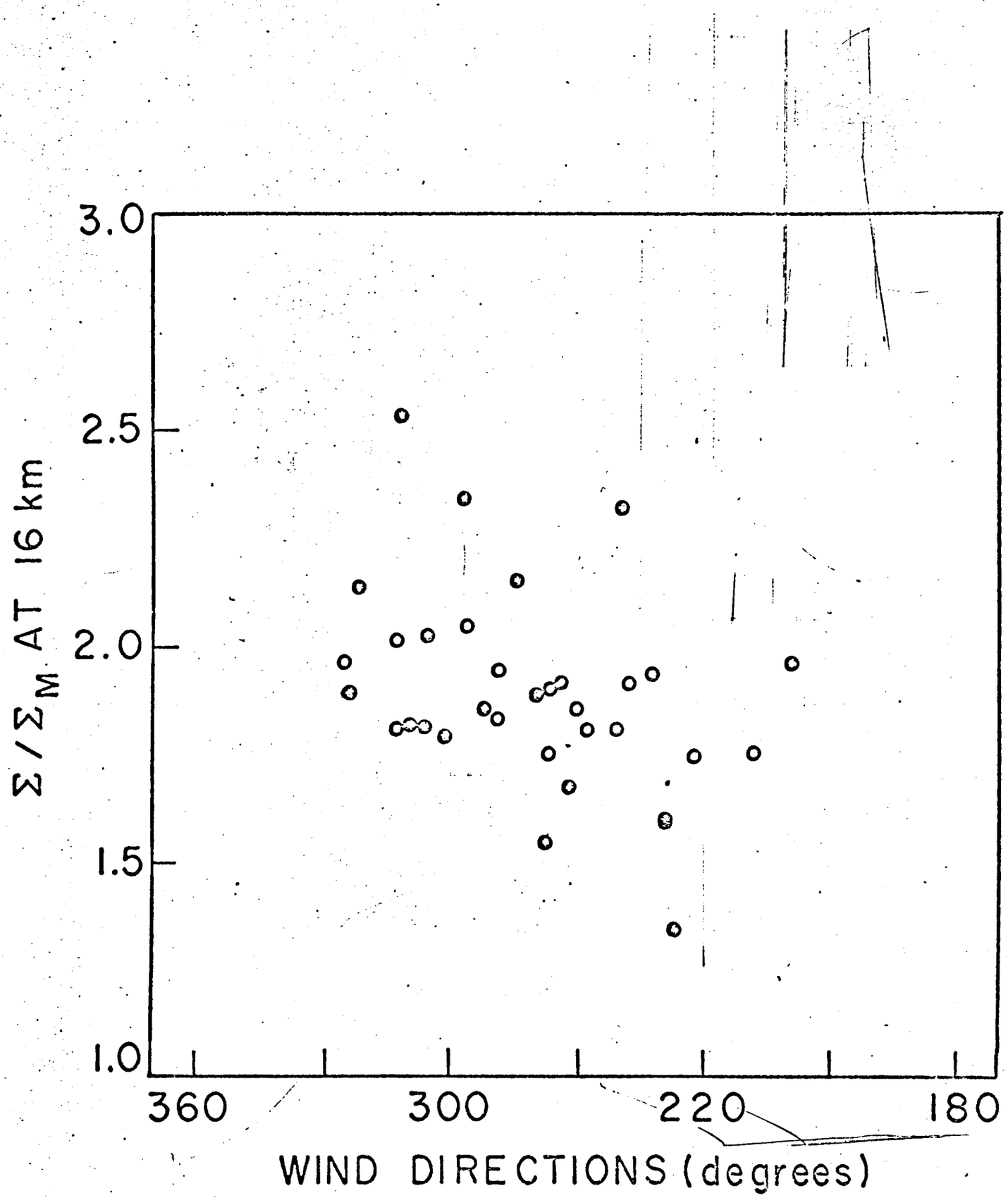

Fig. 15. Relation between the observed scattering ratio Σ/Σ_M at 16 km altitude and the wind direction at 100 mb.

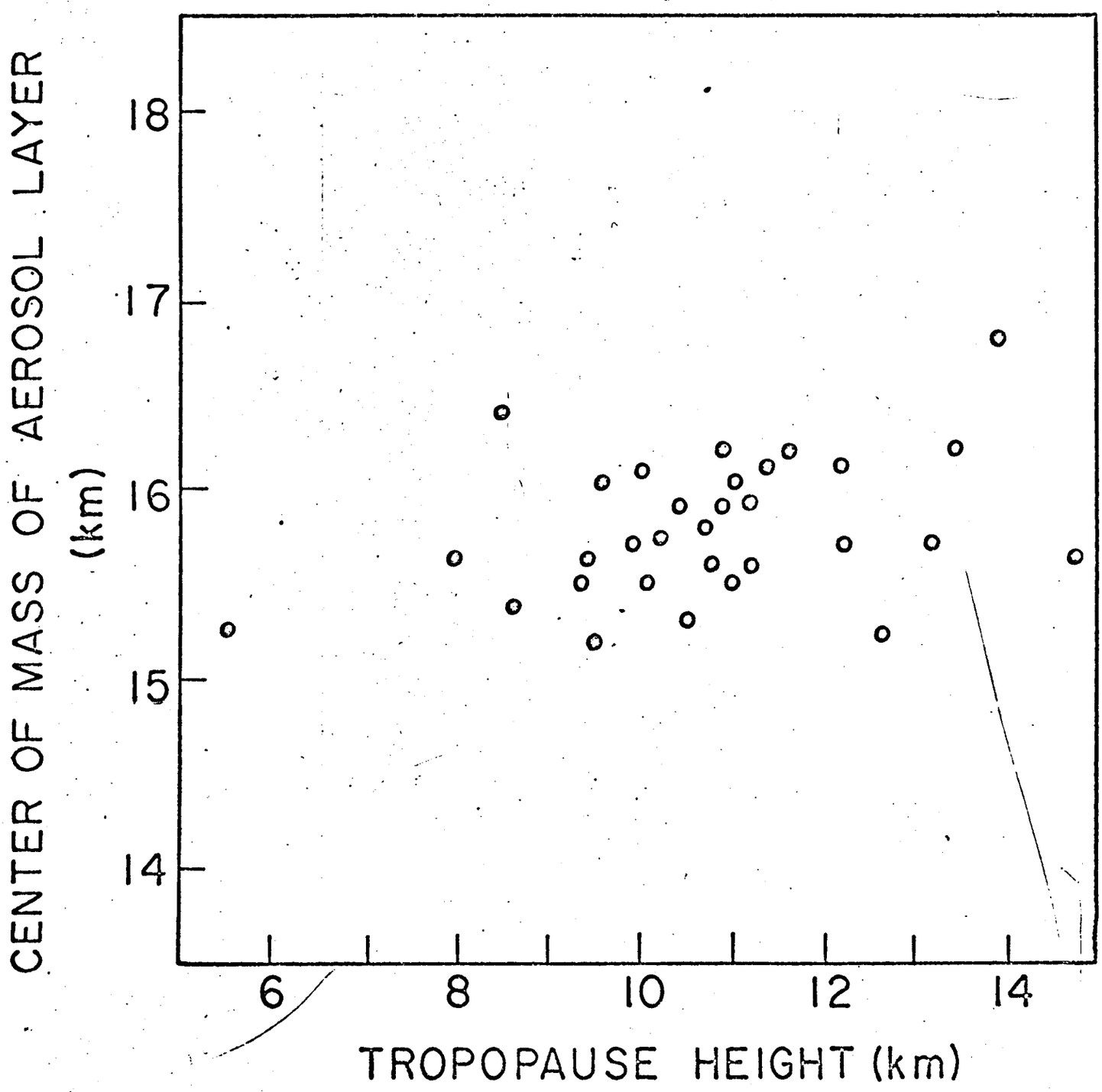

Fig. 16. Relation between the height of the center of mass of the aerosol layer and the height of the tropopause.

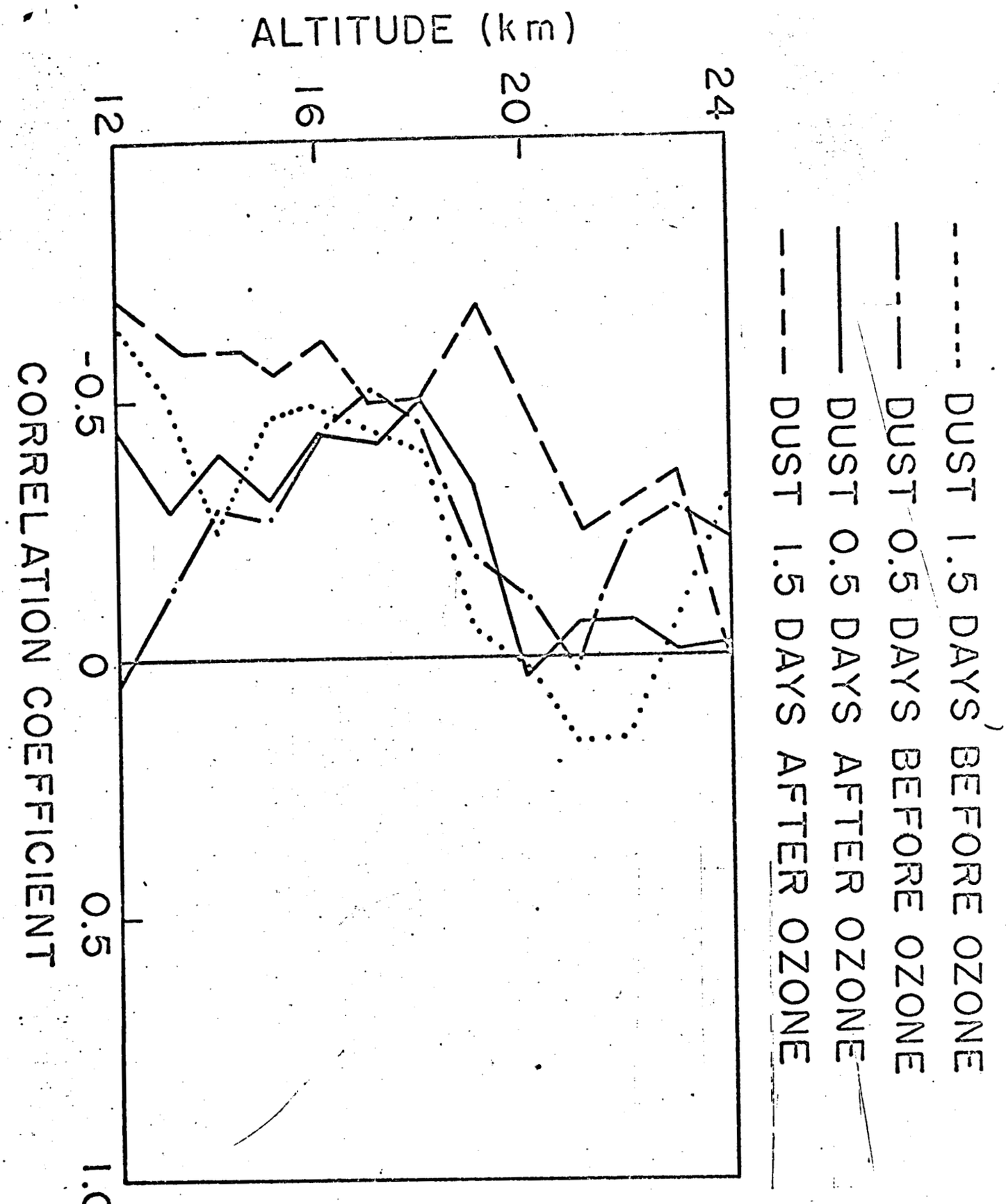

Fig. 17. Correlation coefficients for measurements of the ozone and dust content of the lower stratosphere as a function of altitude for several different time lags between the observations.

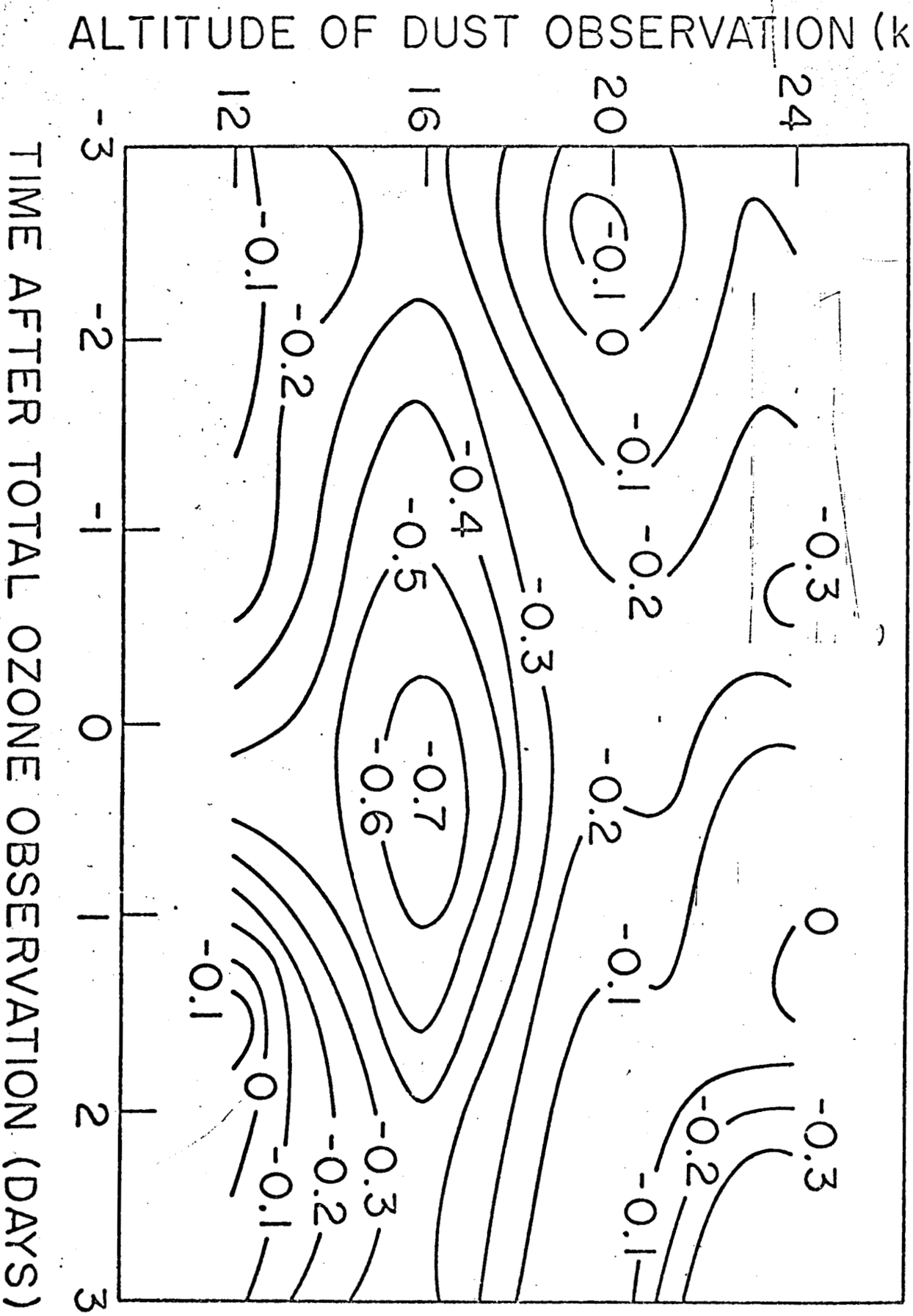

Fig. 18. Correlation coefficients for total ozone and the dust content of the lower stratosphere as a function of the time lag between the observations and the altitude of the dust measurements.

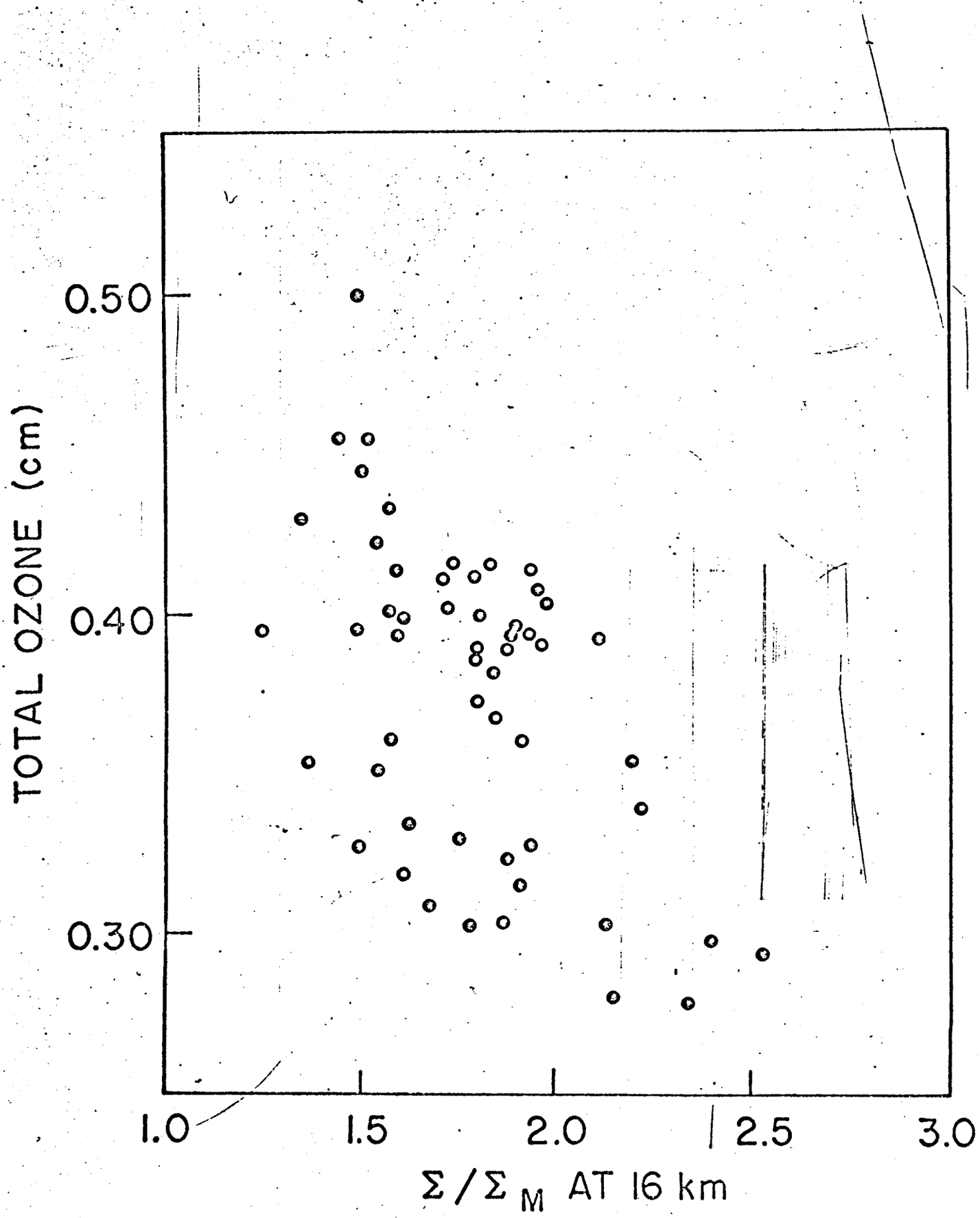

Fig. 19. Relation between the observed scattering ratio Σ/Σ_M at 16 km altitude and the total amount of ozone in the atmosphere.

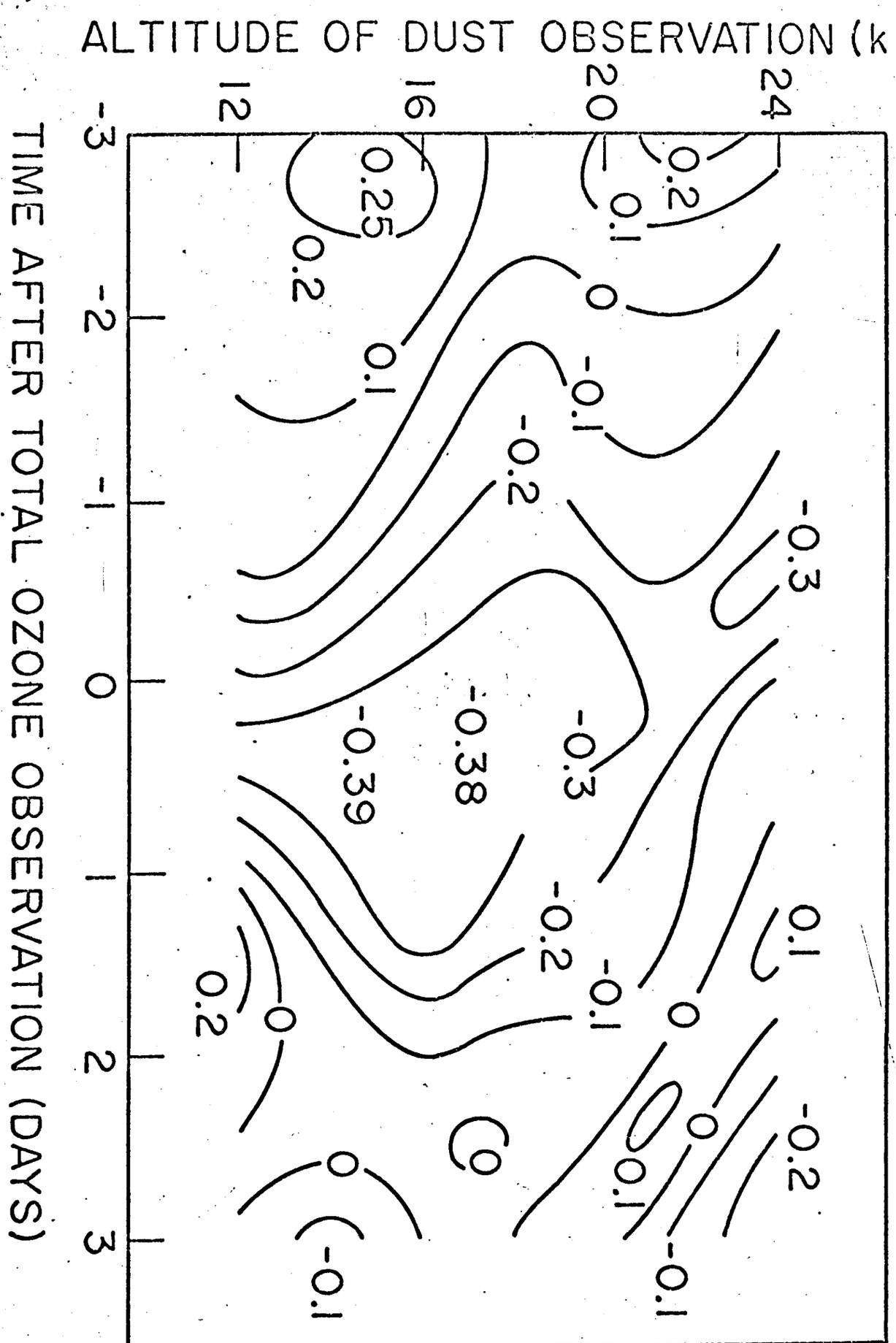

Fig. 20. Correlation coefficients for deviations from the monthly mean of total ozone measurements and the dust content of the lower stratosphere as a function of the time lag between the observations and the altitude of the measured dust.

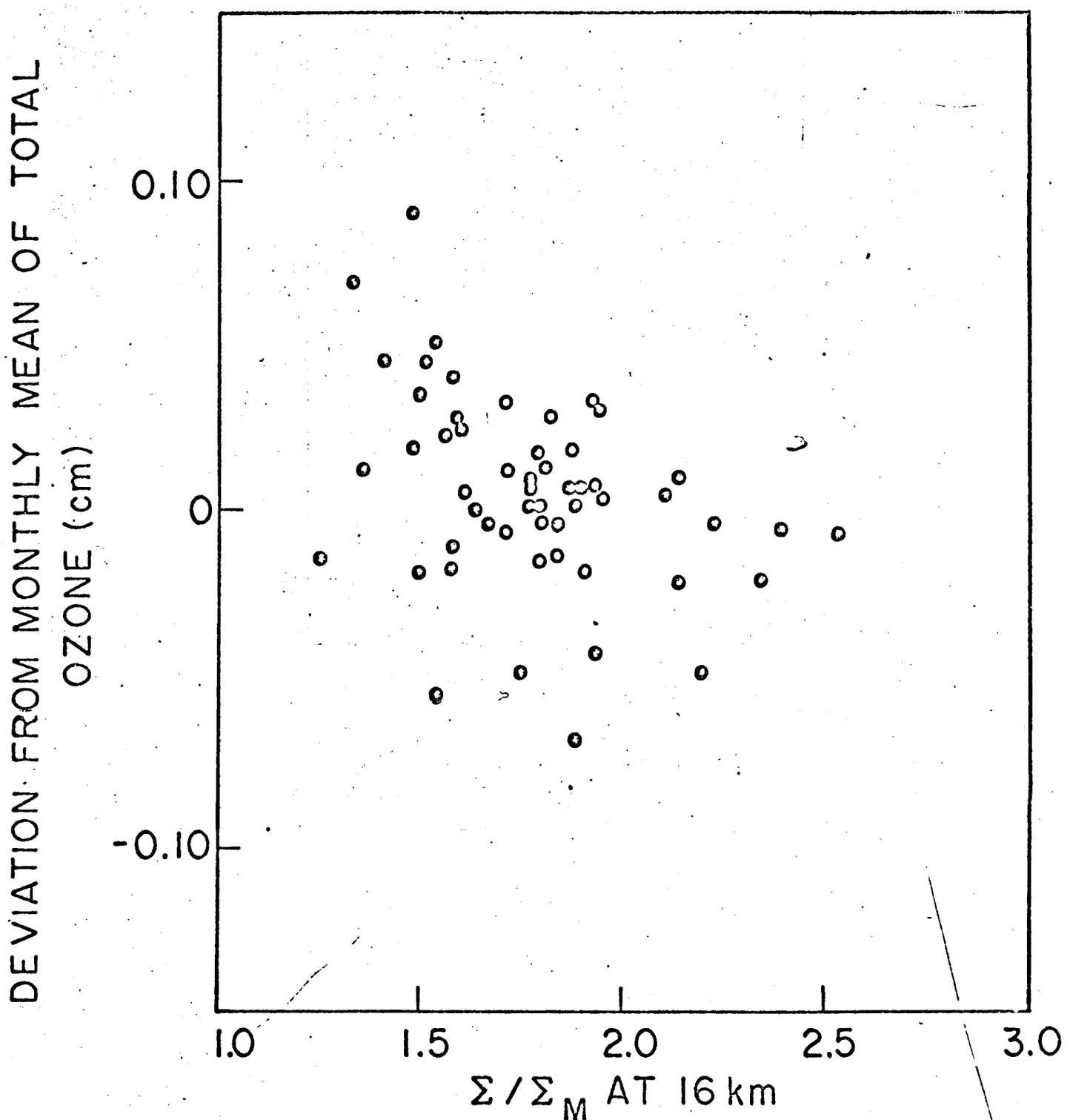

Fig. 2. Relation between the observed scattering ratio Σ/Σ_M at 16 km altitude and the deviations from the monthly mean of the total amount of ozone in the atmosphere.

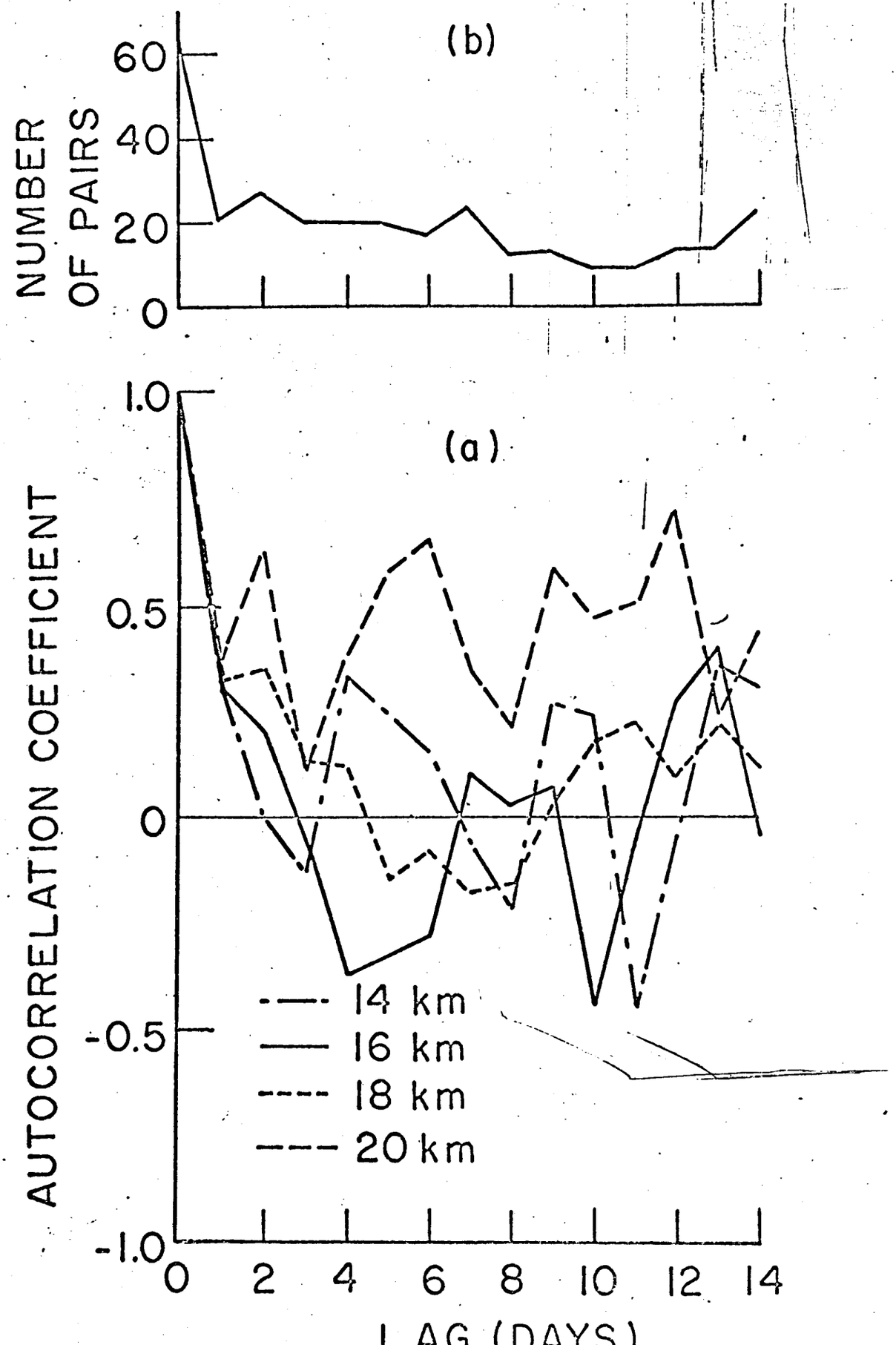

Fig. 22. (a) Autocorrelation coefficients for stratospheric ozone observations near the center of mass of the aerosol layer. (b) Number of data pairs used to compute the coefficients.